



AFRL-RW-EG-TP-2011-024

Matrix Isolation Spectroscopy Applied to Positron Moderation in Cryogenic Solids

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July 2011

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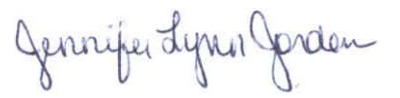
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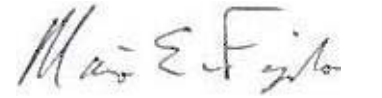
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REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188		
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1. REPORT DATE (DD-MM-YYYY) 07-2011		2. REPORT TYPE Briefing Charts		3. DATES COVERED (From - To) 1 October 2005 – 1 July 2011	
4. TITLE AND SUBTITLE Matrix Isolation Spectroscopy Applied to Positron Moderation in Cryogenic Solids			5a. CONTRACT NUMBER		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER 62602F		
6. AUTHOR(S) Mario E. Fajardo, Christopher D. Molek, C. Michael Lindsay			5d. PROJECT NUMBER 2502		
			5e. TASK NUMBER 10		
			5f. WORK UNIT NUMBER 15		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Air Force Research Laboratory Munitions Directorate Ordnance Division Energetic Materials Branch Eglin AFB, FL 32542-5910			8. PERFORMING ORGANIZATION REPORT NUMBER AFRL-RW-EG-TP-2011-024		
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) Air Force Research Laboratory, Munitions Directorate Ordnance Division Energetic Materials Branch (AFRL/RWME) Eglin AFB FL 32542-5910 Technical Advisor: Dr. Jennifer L. Jordan			10. SPONSOR/MONITOR'S ACRONYM(S) AFRL-RW-EG		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-RW-EG-TP-2011-024		
12. DISTRIBUTION / AVAILABILITY STATEMENT Distribution A: Approved for public release; distribution unlimited. Approval Confirmation 96 ABW/PA # 96ABW-2011-0324, dated June 2, 2011					
13. SUPPLEMENTARY NOTES DISTRIBUTION STATEMENT INDICATING AUTHORIZED ACCESS IS ON THE COVER PAGE AND BLOCK 12 OF THIS FORM.					
14. ABSTRACT We report results of Matrix Isolation Spectroscopy (MIS) experiments performed on working cryogenic rare gas solid (RGS) positron (e^+) moderators. The e^+ is the antiparticle to the electron, ¹⁻³ and positrons are produced by energetic processes that result in very broad kinetic energy distributions (KEDs) - typically spanning hundreds of thousands of electron volts (eV). Trapping and manipulating e^+ with electromagnetic fields requires narrowing these KEDs below a few eV, which is accomplished <i>via</i> velocity-dependent interactions in a normal matter "moderator." A fast e^+ entering a wide bandgap dielectric RGS moderator slows rapidly within the first ~ picosecond, producing a track of ionized and electronically excited species. However, once KE_{e^+} drops below the ~ 10 eV minimum required for generating such excitations, these interactions abruptly switch off, and the e^+ enters a phase of "hyperthermal diffusion" that can last for nanoseconds. Positrons that reach a free surface of the moderator before annihilating with an electron may escape into vacuum where they can be manipulated. The best known e^+ moderator is cryogenic solid Ne, which still only delivers efficiencies < 1 %; the other 99+ % of the nascent fast e^+ are wasted. Additionally, the RGS moderator efficiency is known to decrease during operation, which is attributed variously to the buildup of radiation damage, and/or to contamination of the moderator surface by residual gas deposition. We constructed a novel apparatus that permits optical access to a working cryogenic solid moderator. Our original motivation was to test our hypothesis that solid parahydrogen (pH_2) should be an even better e^+ moderator than solid Ne, while simultaneously monitoring the condition of the moderators by infrared (IR) absorption spectroscopy. Unfortunately, the performance of our ortho/para hydrogen converter (o/p converter) has deteriorated and we only achieved residual orthohydrogen (oH_2) concentrations ~ 1000 ppm in these experiments. While we successfully observed the production of slow e^+ from Ne, Ar, and Kr moderators (background corrected count rates ~ 10 Hz), our oH_2/pH_2 experiments failed to yield a statistically significant slow e^+ signal. Indeed, a ~ 1 μ m thick oH_2/pH_2 overcoat was sufficient to kill the slow e^+ signal from a working solid Ne moderator. We performed several experiments with deliberately doped (CO and H_2O) RGS moderators, and with RGS moderators overcoated with solid CO and water ice, in which we measured quantitatively the dopant concentrations and the thicknesses of the moderator and overcoat layers from the IR spectroscopic data. Our results indicate that the impact of H_2O impurities on RGS moderator efficiency has been overestimated in the literature, and point instead to residual H_2 gas as a possible culprit.					
15. SUBJECT TERMS Positron moderation, rare gas solid, matrix isolation, parahydrogen					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE	UL	37	Mario E. Fajardo
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Munitions Directorate, AFRL/RWME

MATRIX2011
Physics & Chemistry of Matrix Isolated Species
U. British Columbia, Vancouver, Canada, 10-15 Jul 11



Outline



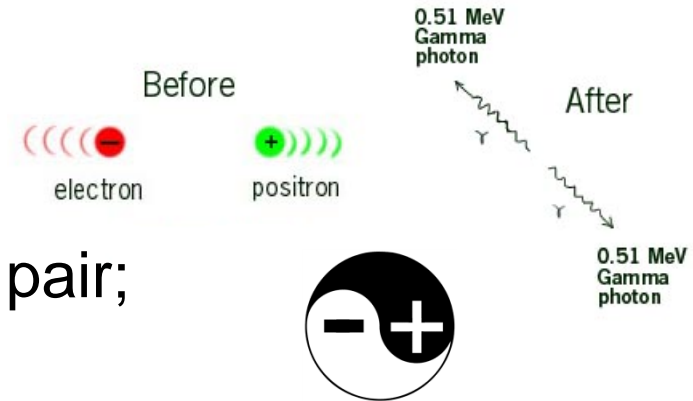
- Positrons introduction
- Reality check: near- vs. far-term applications
- Positron production, moderation, and trapping
- Solid parahydrogen (pH_2) as a positron moderator
- Slow electron transport experiments & simulations
- Positron moderation experiments
 - solid Ne moderator
 - solid pH_2 “moderator”
 - H_2O -doped solid Kr moderator
 - H_2O -ice coated Kr moderator
- Summary & Future Directions



Introduction - Positrons



- The positron (e^+) is the antiparticle to the electron (e^-).
- Positrons are stable, but annihilate with electrons to produce γ -rays.
- “Positronium” (Ps) is a bound e^+/e^- pair; analogous to the H-atom (p^+/e^-).
- Sum of γ -ray energies: $E \approx 2 m_e c^2 \approx 1.02 \text{ MeV}$;
 \Rightarrow no photonuclear reactions or secondary radioactivity, but lots of energy/mass (**$>10^{10}$ x chemical energies**).
- However, bulk positron or positronium production and storage are problematic (understatement!).

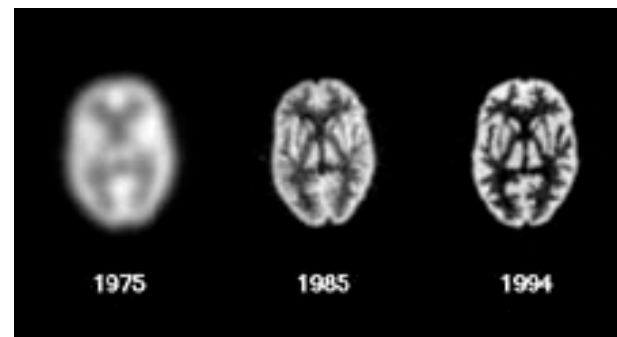
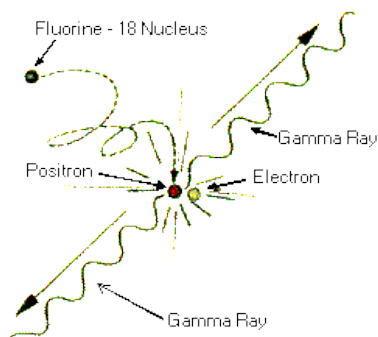




Current Positron Applications



- 2- γ decay exploited in Positron Emission Tomography (PET) scanners.



- Positrons localize & annihilate preferentially at defects in materials. Basis for materials diagnostics:
Doppler Broadening of Annihilation Radiation (DBAR)
& Positron Annihilation Lifetime Spectroscopy (PALS).

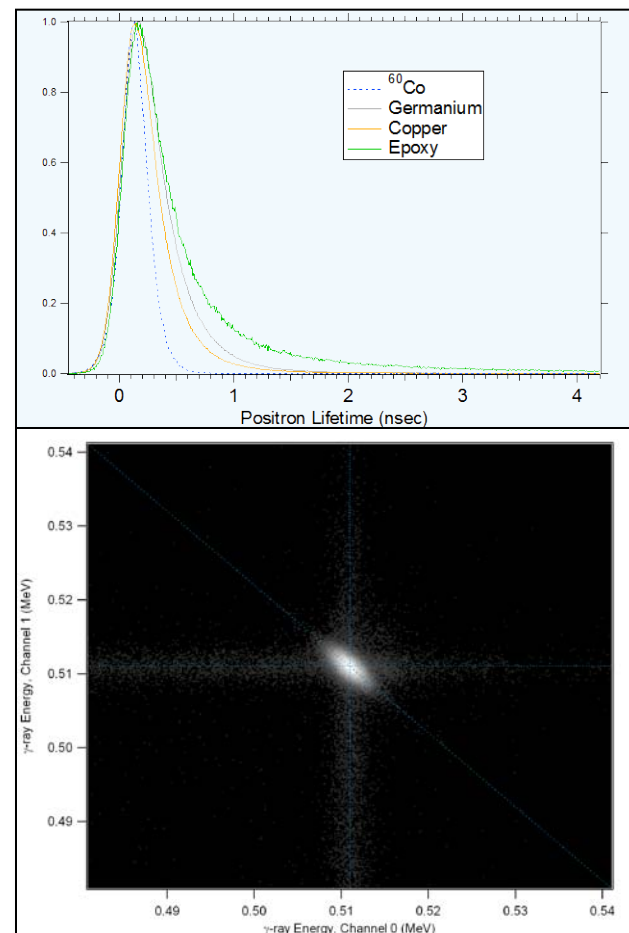
Antimatter is real, and is utilized in hundreds of labs worldwide.



Energetic Materials Characterization



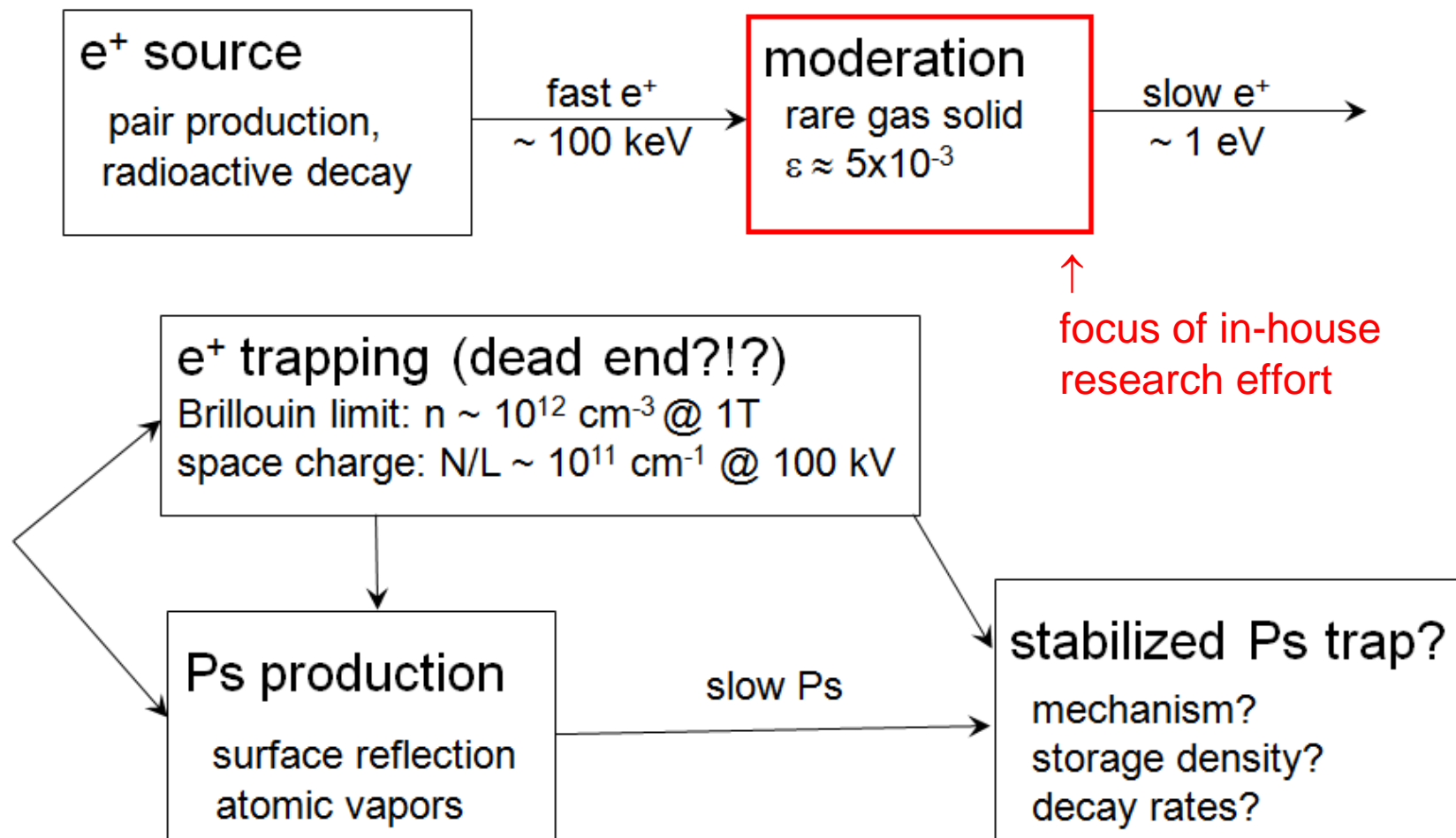
- Voids and defects play critical roles in energetic materials (EM).
- Few data available on nanoscale (sub-micron) defects.
- In-house NanoEnergetics effort underway to develop characterization tools to probe nanoscale defects in EM.
- Objective: measure defect size and concentration in EM and their relationship to processing and mechanical insults, and EM sensitivity.
- Accomplishments: PALS and 2D-DBAR instrumentation in use (home-built, pushing state-of-the-art).
- POC: Dr. C.M. Lindsay (ARFL/RWME).



Near-term payoffs from in-house positron investments.



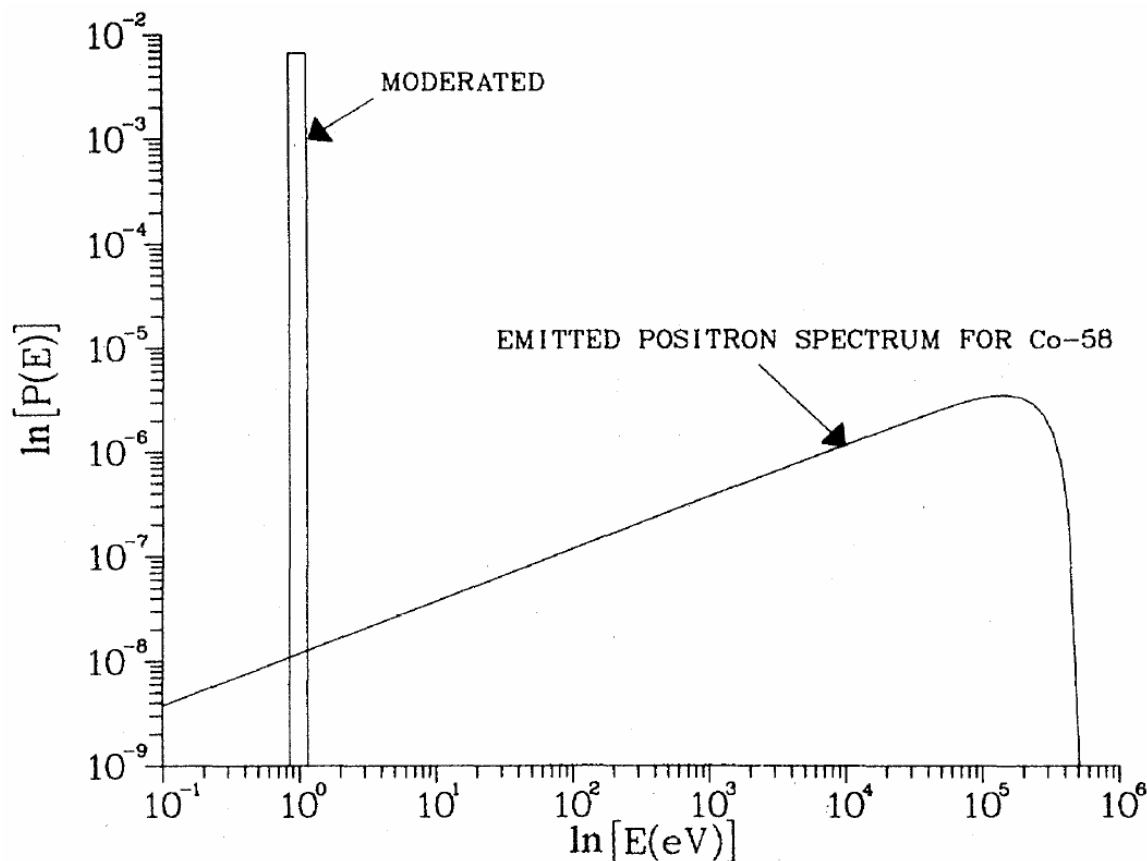
Generic Positron Trapping Scheme



Moderation efficiency improvements are cumulative for slow e⁺ applications.



Moderation vs. Velocity Selection

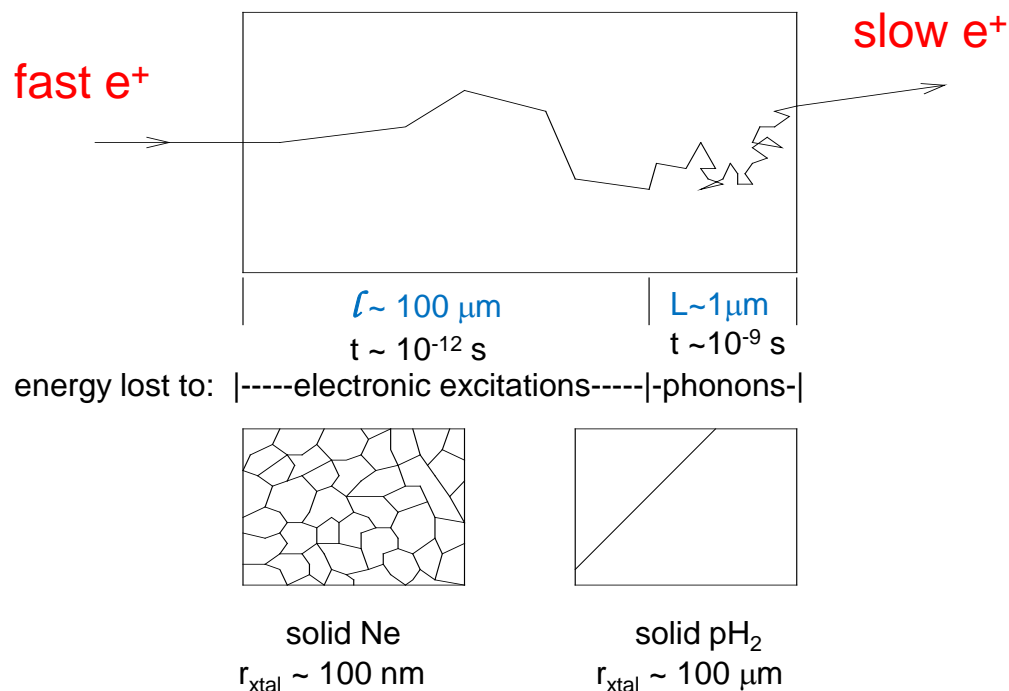


P.J. Schultz and K.G. Lynn, Rev. Mod. Phys. **60**, 701 (1988).

Moderation converts fast positrons into slow positrons.



Positron Moderation Physics



E.M. Gullikson and A.P. Mills, Jr.,
Phys. Rev. Lett. **57**, 376 (1986).

- Fast positrons decelerate rapidly in the first \sim picosecond by creation of electronic excitations.
- Slow positrons (below bandgap energy) can only create phonons and diffuse randomly for nanoseconds.
- Positrons which reach a moderator surface can be extracted & recovered.
- Slow positrons scatter, trap and annihilate at defects, becoming lost to the moderation process.
- Conjecture: higher quality RVD pH_2 solids should permit more slow positrons to emerge from deeper within the moderator.

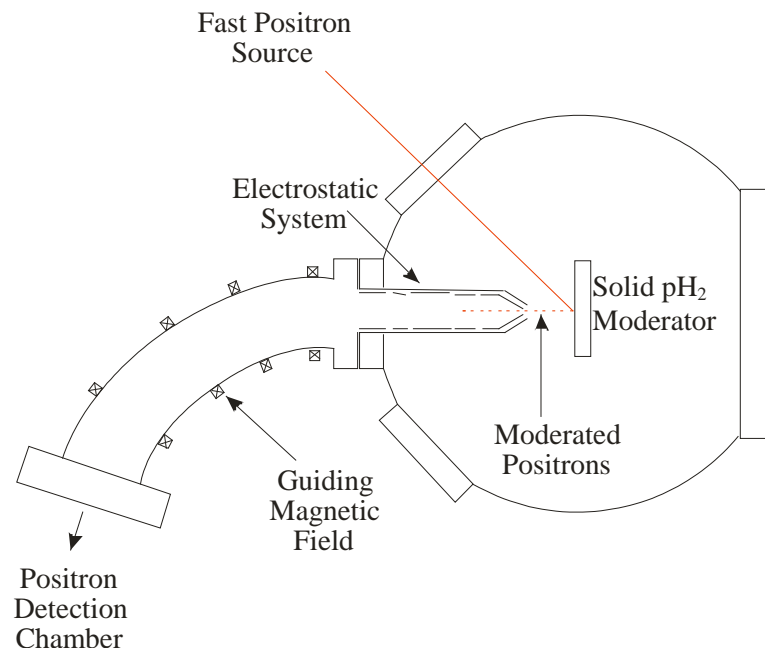
Moderation efficiency increases as $\sim L / \ell$.



Moderation Objectives & Approach



- Develop improved efficiency (ε) positron moderators based on cryogenic ($T \approx 2$ K) RVD pH_2 solids
goal: **10x** improvement over the state of the art (Ne), yielding $\varepsilon \approx 5$ %.
- Determine preferred pH_2 moderator structures and moderator failure modes by *in-situ* spectroscopic characterization.
- Moderate fast positrons from ~ 1 mCi ^{22}Na source in reflection geometry.
- Separate slow from fast positrons using electrostatic and magnetic guiding fields around bend into annihilation chamber.
- Slow positron detection: charged particle and annihilation γ -radiation.



Notional Experimental Diagram

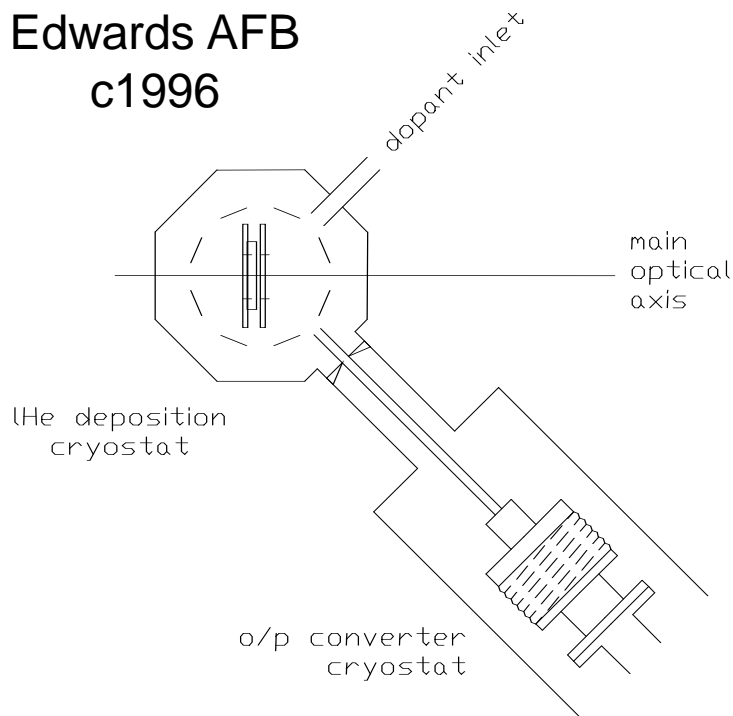
C.D. Molek, "Moderation of Fast Positrons Using Solid Parahydrogen,"
NRC Research Proposal (2007).



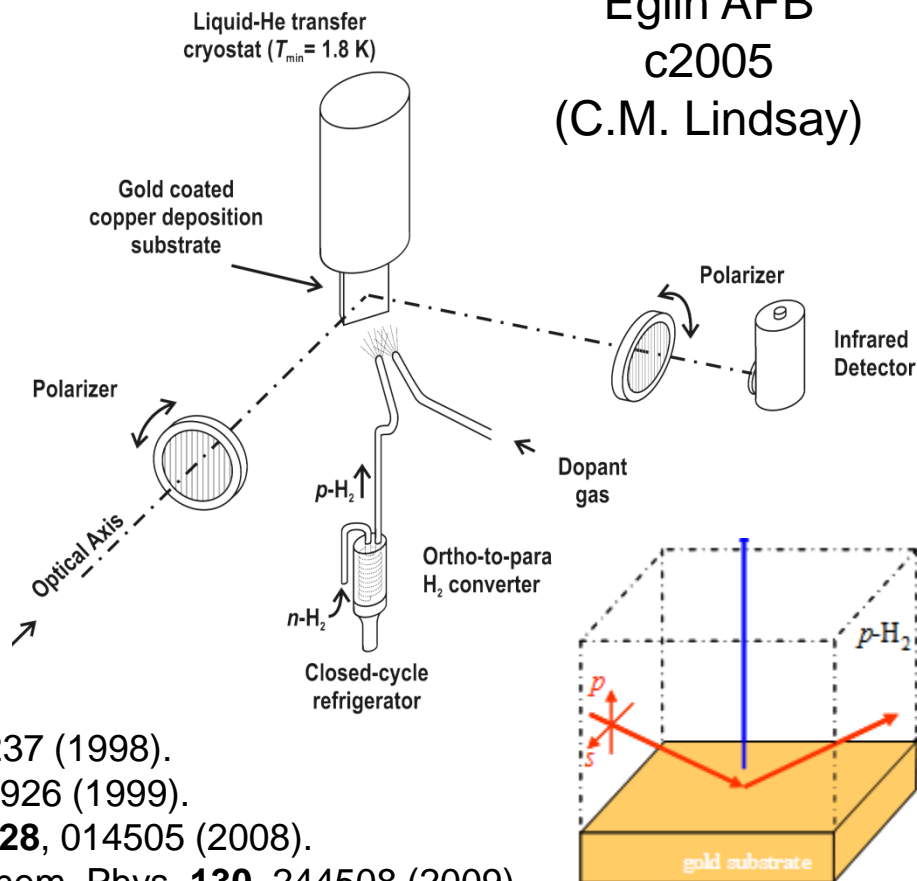
Rapid Vapor Deposition of Solid $p\text{H}_2$



Edwards AFB
c1996



Eglin AFB
c2005
(C.M. Lindsay)

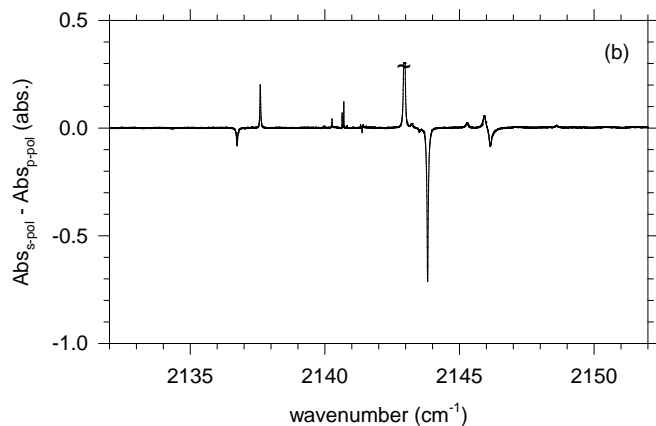
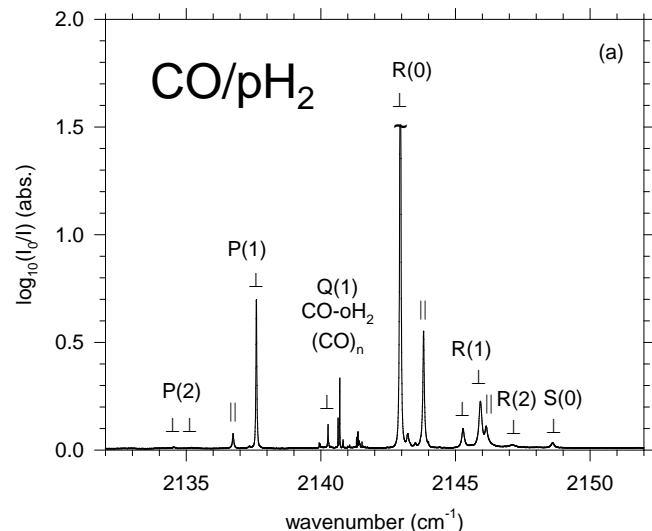


M.E. Fajardo and S. Tam, J. Chem. Phys. **108**, 4237 (1998).
S. Tam and M.E. Fajardo, Rev. Sci. Instrum. **70**, 1926 (1999).
M.E. Fajardo and C.M. Lindsay, J. Chem. Phys. **128**, 014505 (2008).
M.E. Fajardo, C.M. Lindsay, and T. Momose, J. Chem. Phys. **130**, 244508 (2009).

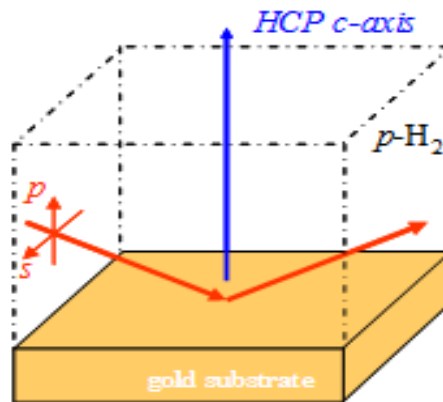
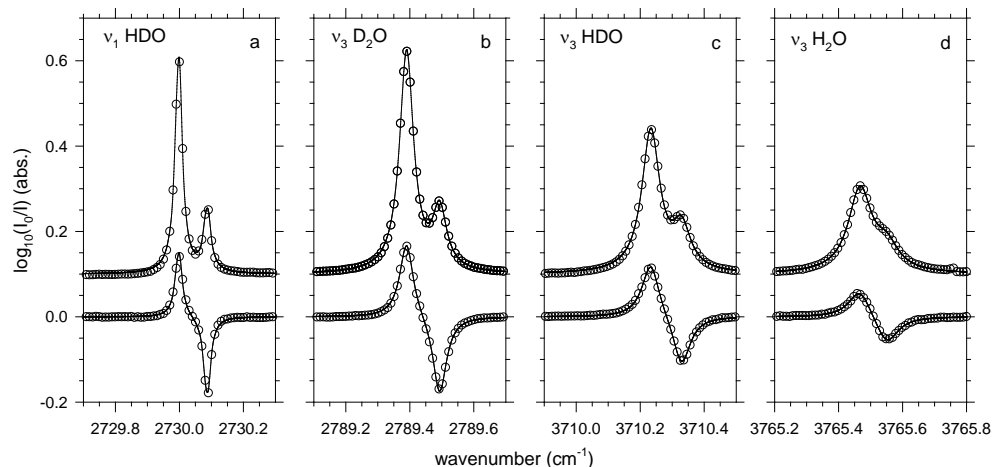
Building upon solid $p\text{H}_2$ expertise developed during 1990's "HEDM" program.



Polarized IR Absorption Spectra



water/pH₂



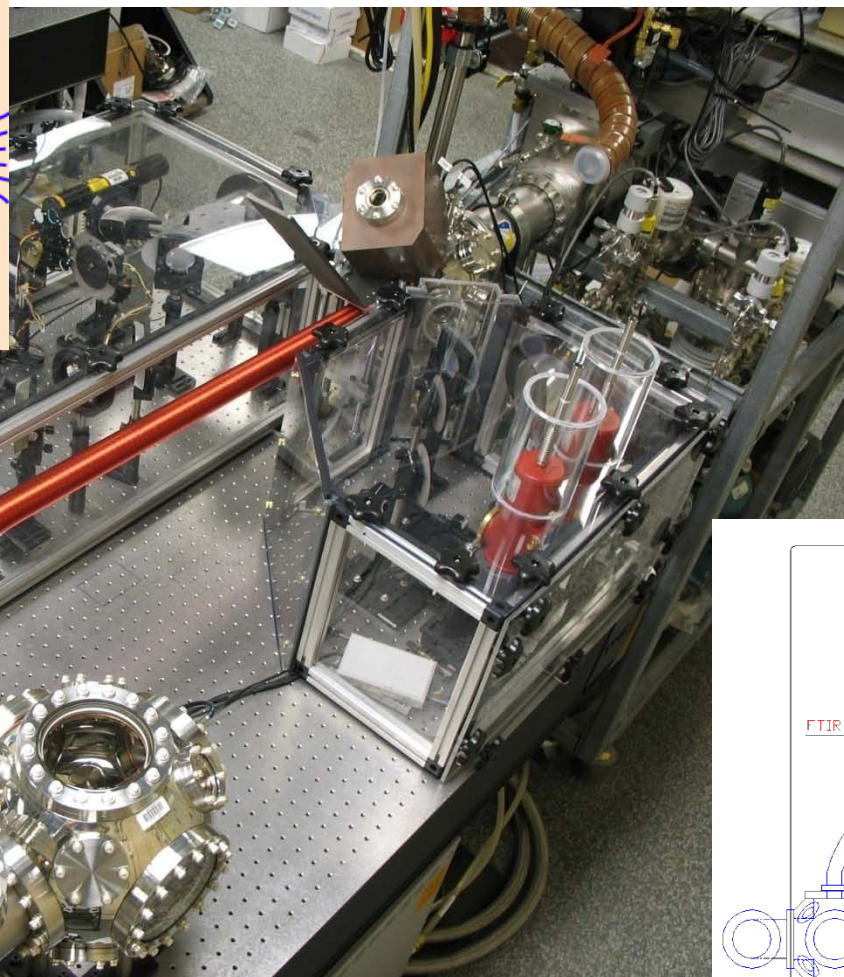
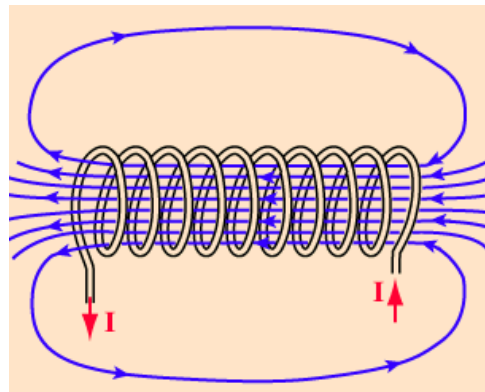
Polarization dependence as predicted by Crystal Field Theory modeling.

M.E. Fajardo and C.M. Lindsay, J. Chem. Phys. **128**, 014505 (2008).
M.E. Fajardo, C.M. Lindsay, and T. Momose, J. Chem. Phys. **130**, 244508 (2009).

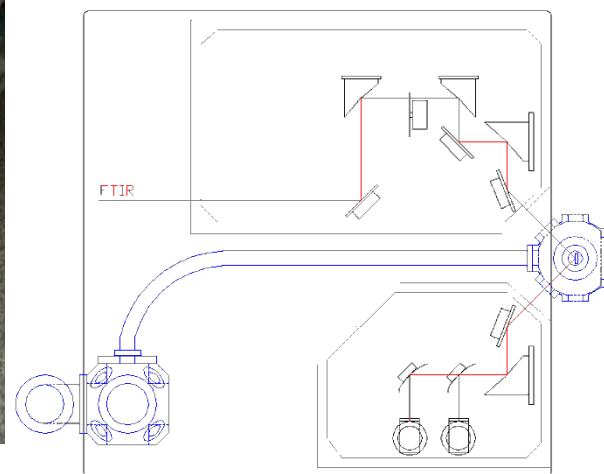
Annealed hcp c-axes all parallel to substrate surface normal! Single crystal?



Preliminary Electron Transport Expt.

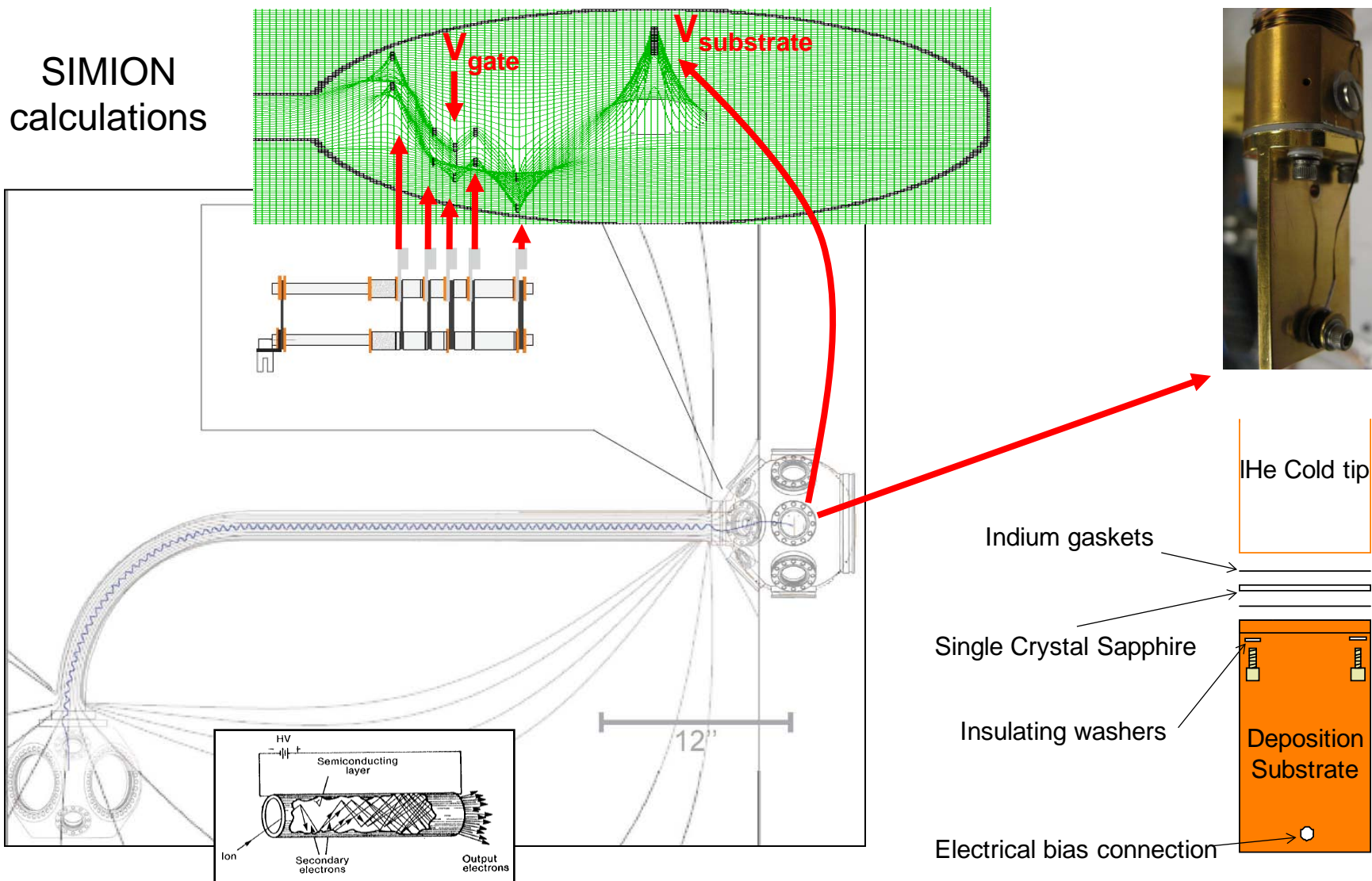


Shown configured for initial slow electron transport experiments through bent solenoid using hot-filament electron source and Faraday cup detector.





Ion Optics & Deposition Substrate



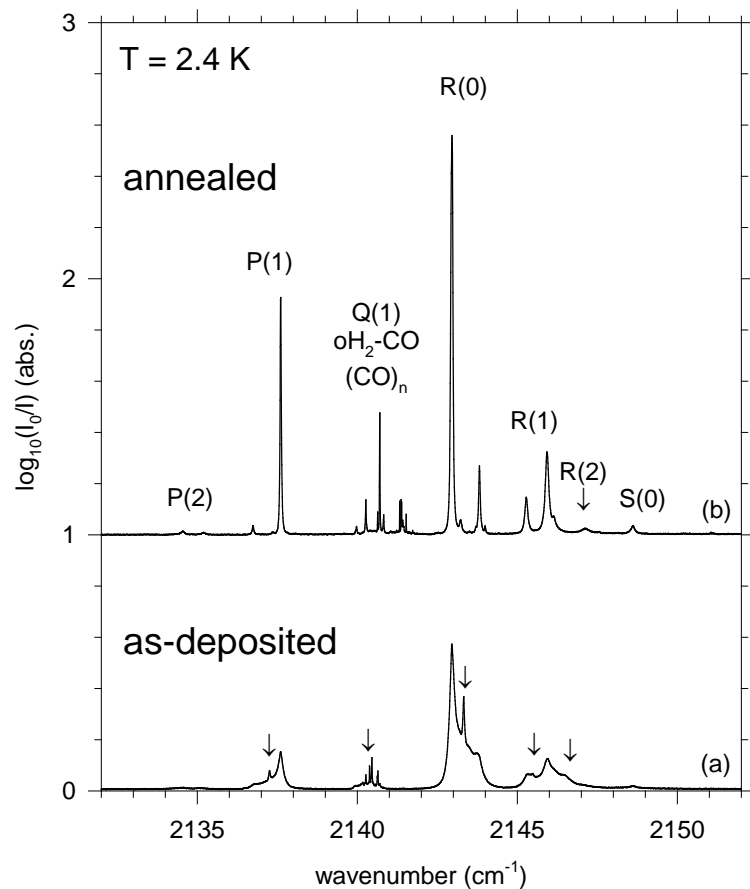
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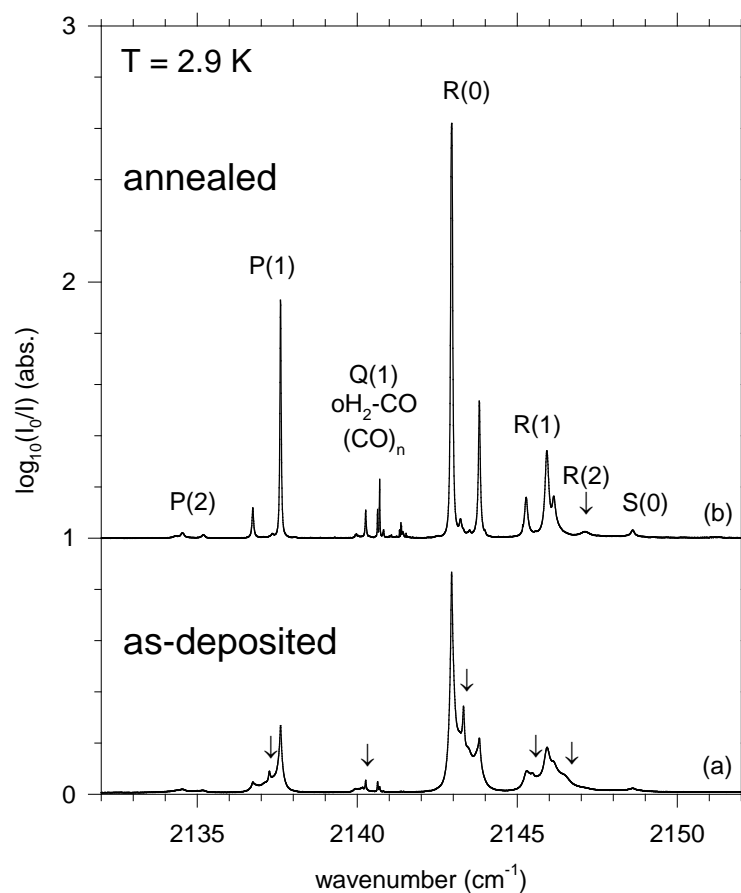
Isolated Deposition Substrate



CO/pH₂ before modification



CO/pH₂ after modification



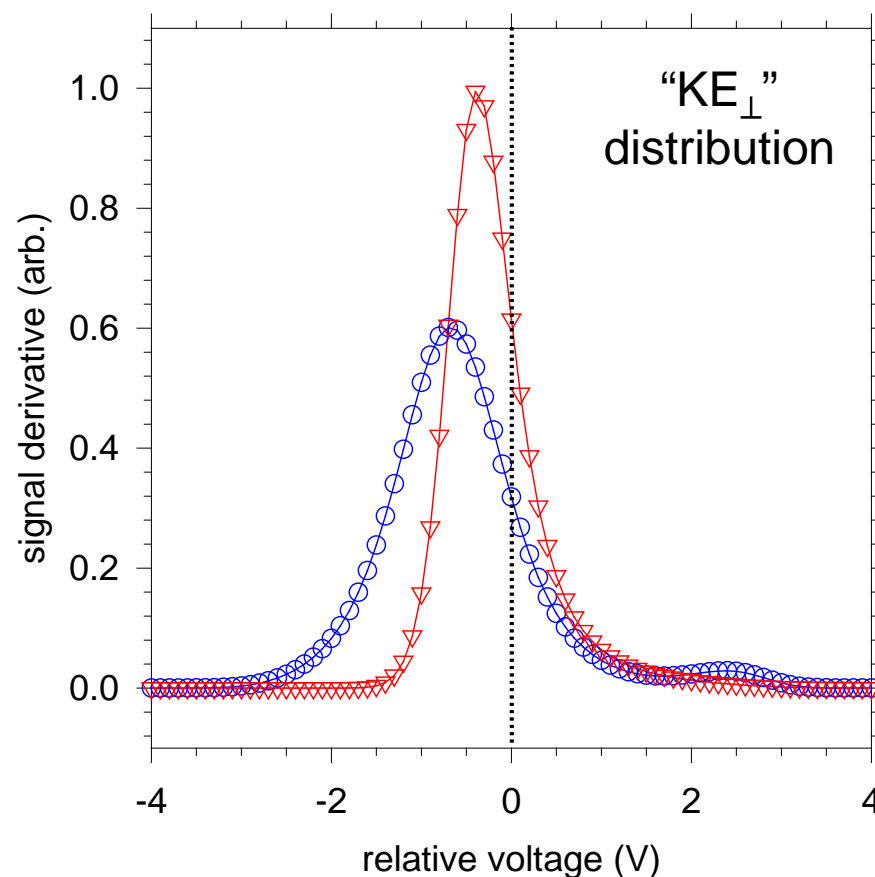
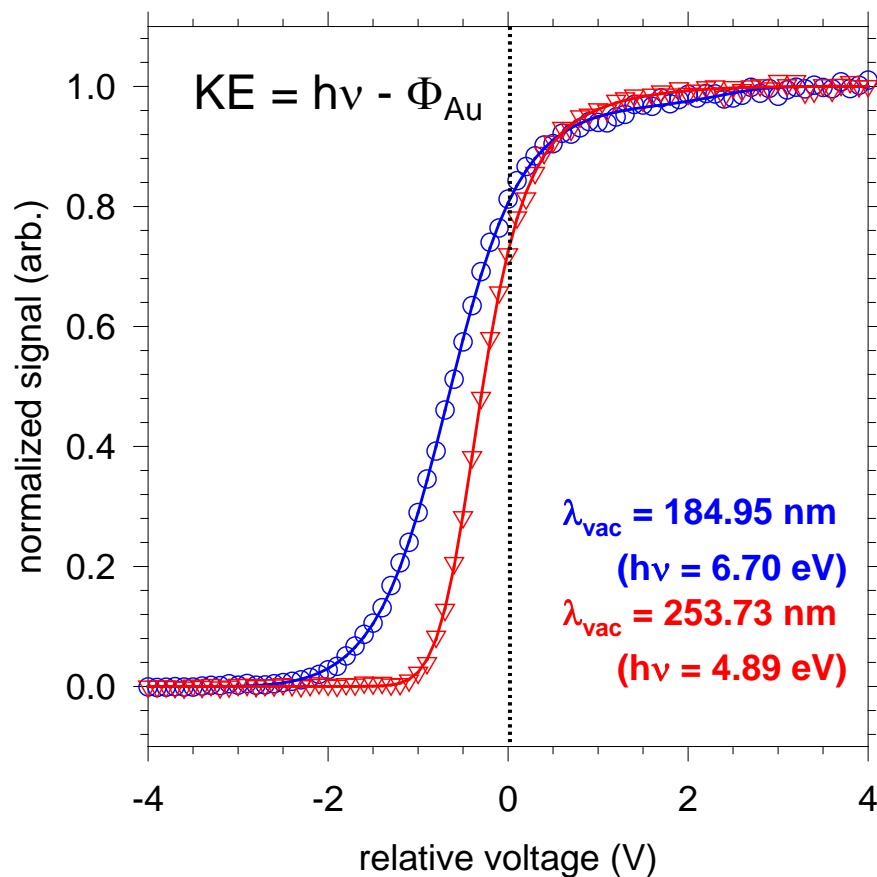
Only minor changes to spectra from ≈ 0.5 K higher substrate base temperature.



Photoelectron Transport Expts.



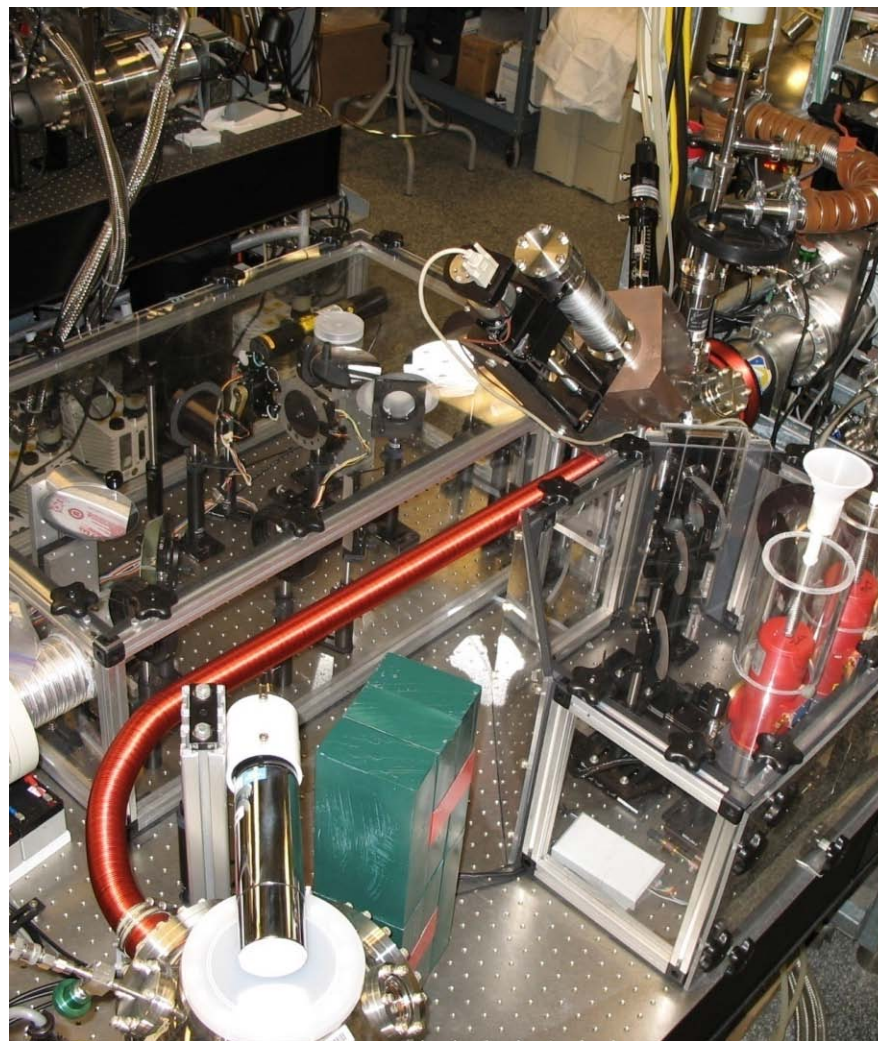
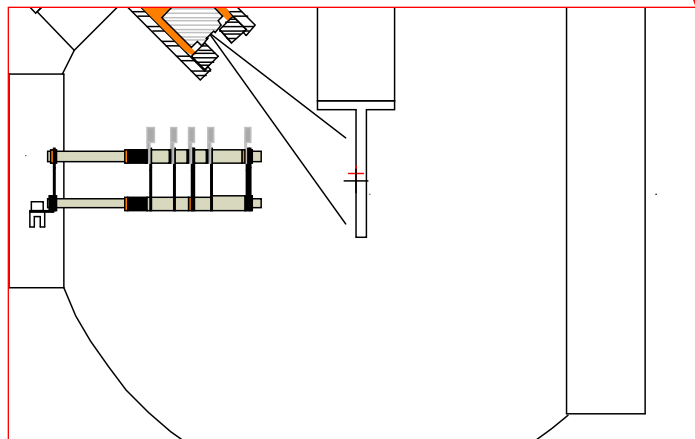
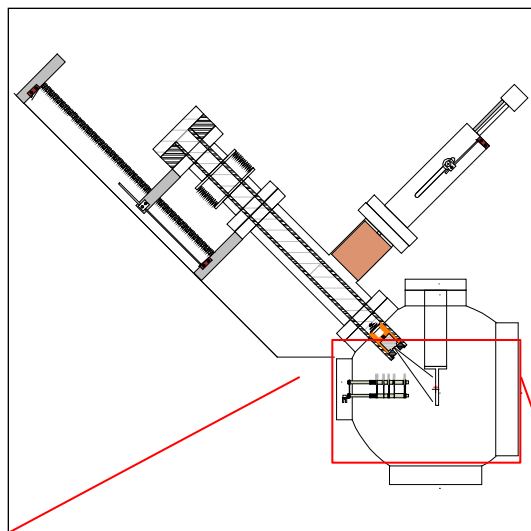
Hg/Ar pen lamp + UV bandpass filters + pinhole apertures



Electron throughput and energy resolution acceptable for e^+ moderation expts.



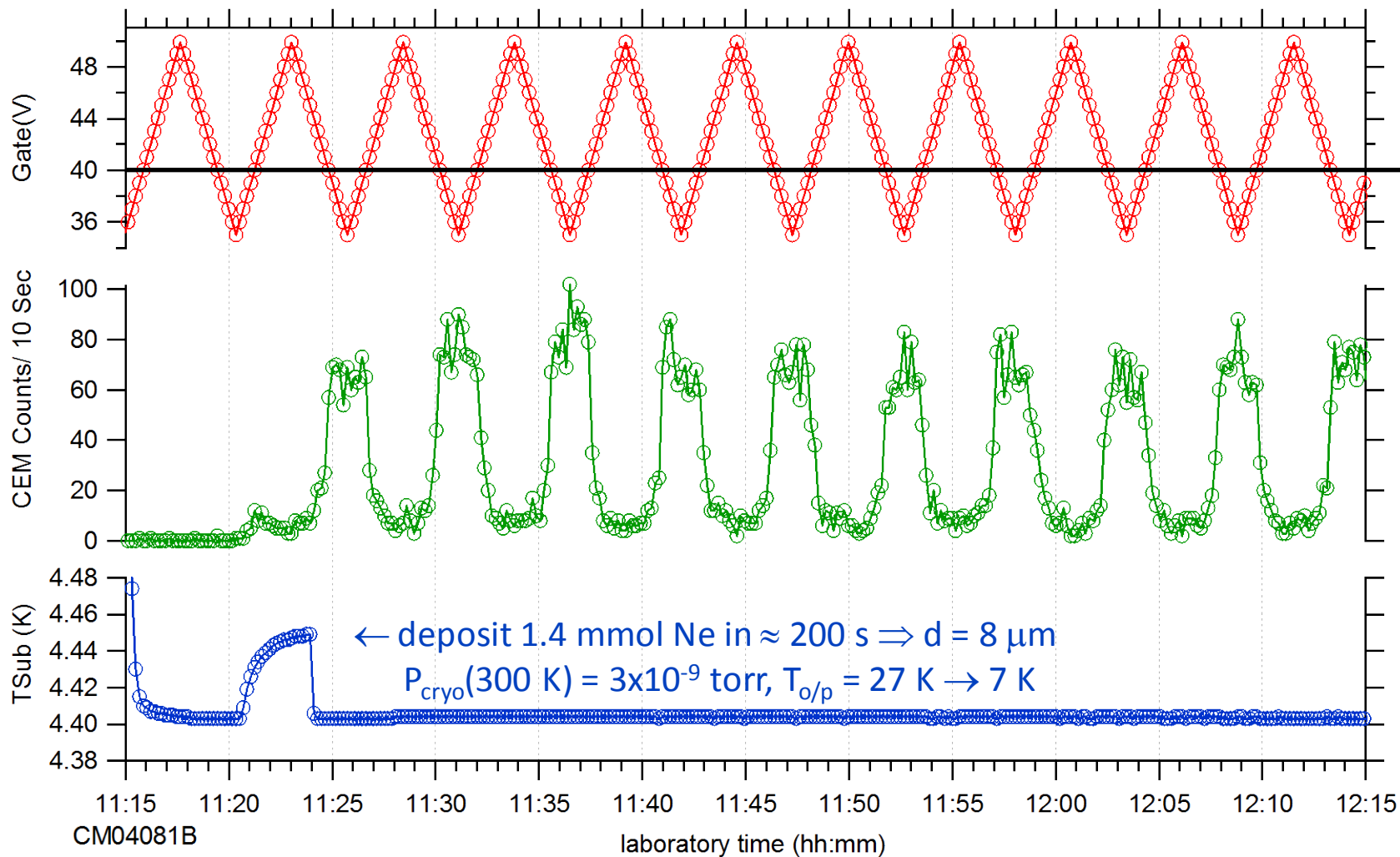
^{22}Na Positron Source – 1 mCi



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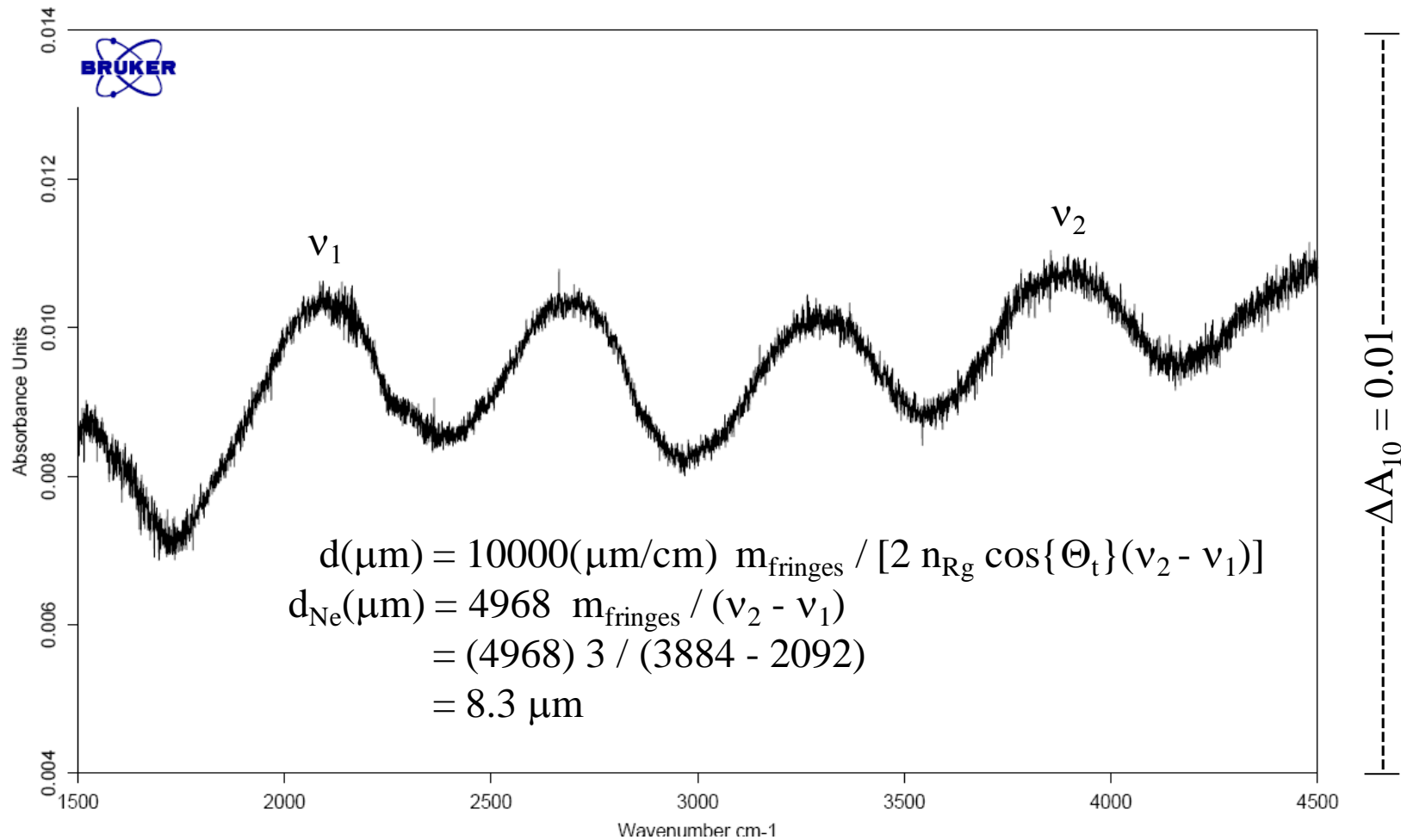


Ne Moderator Deposition



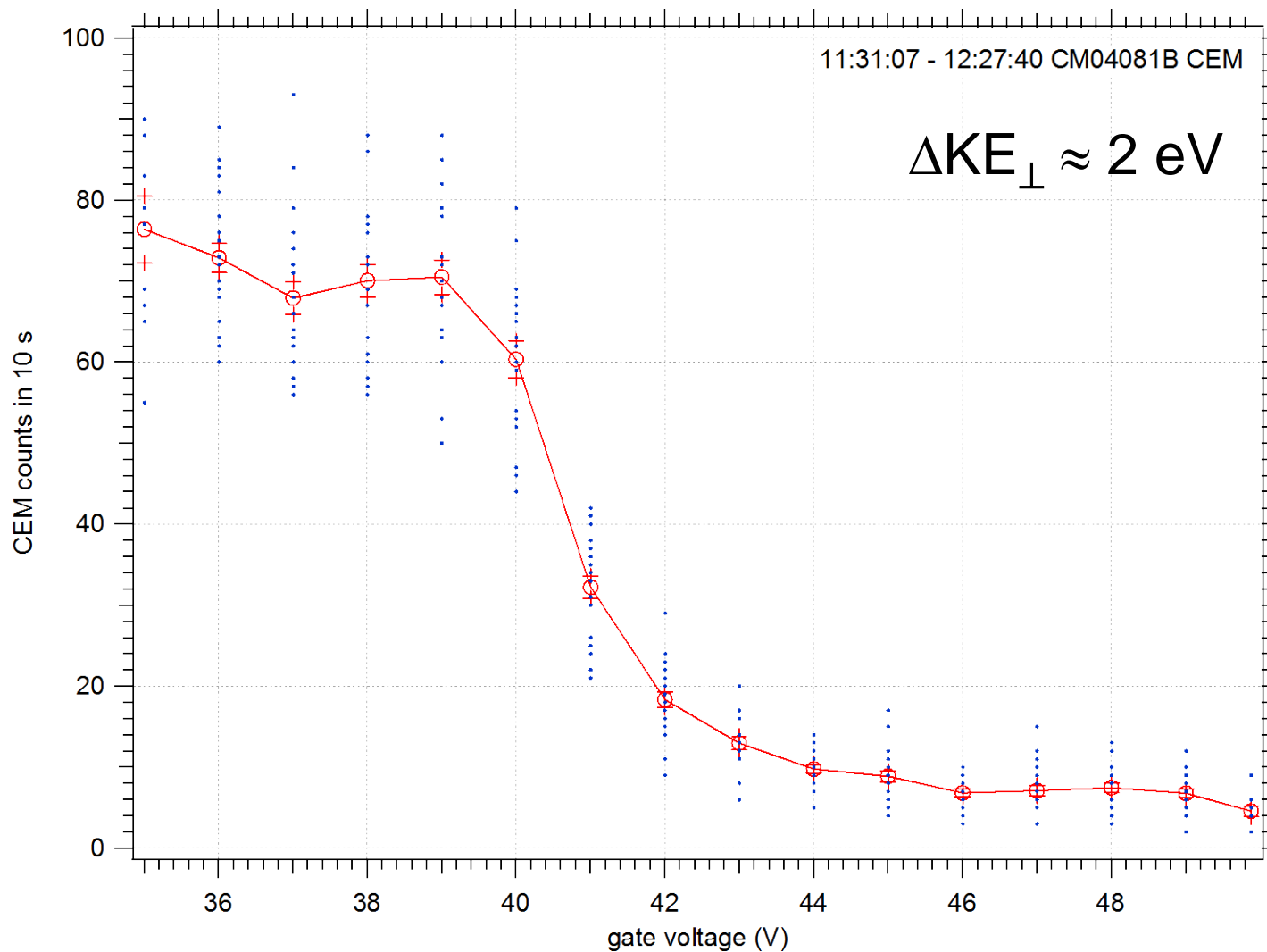


Ne Moderator Thickness



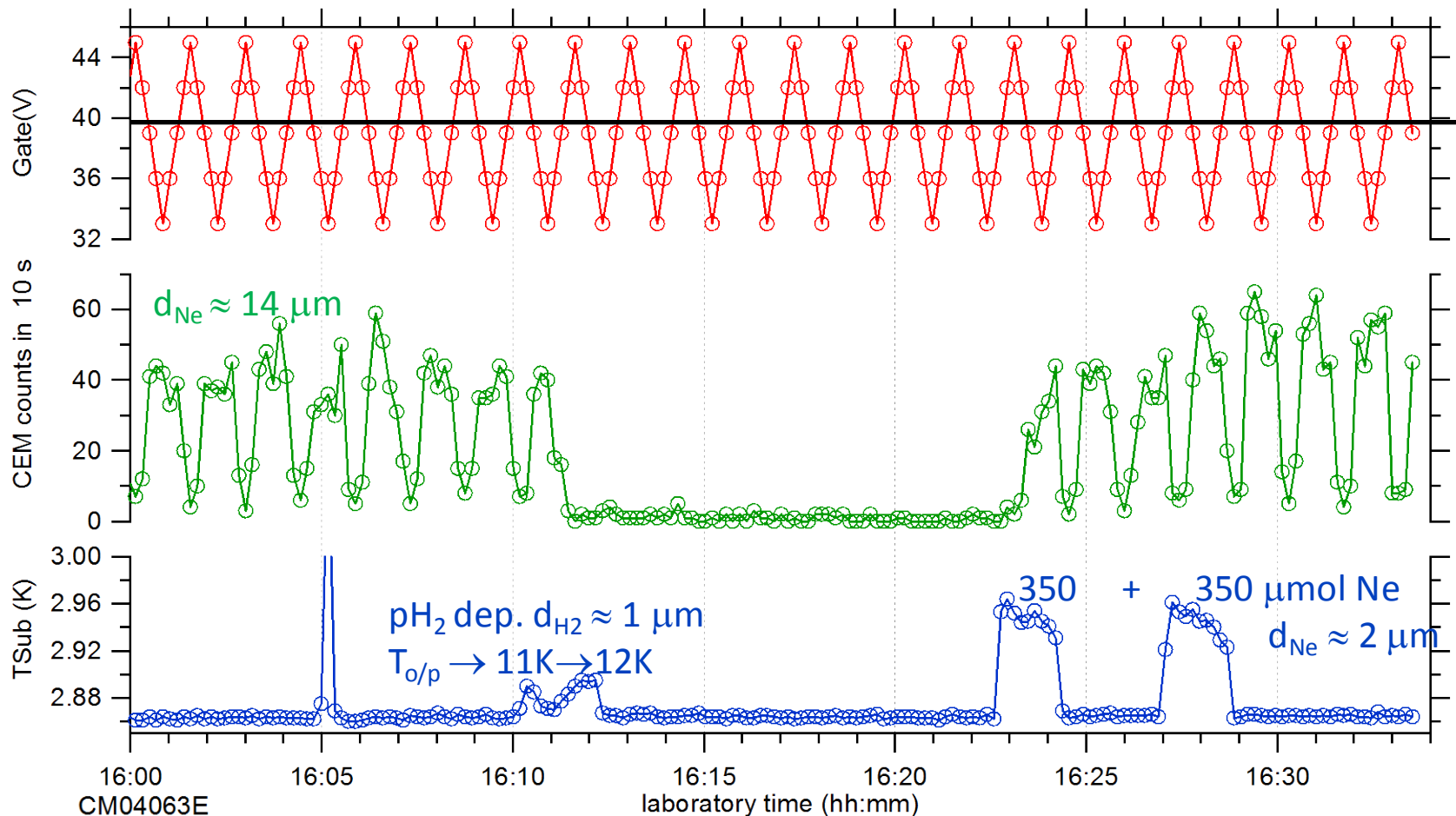


Ne Moderator Performance





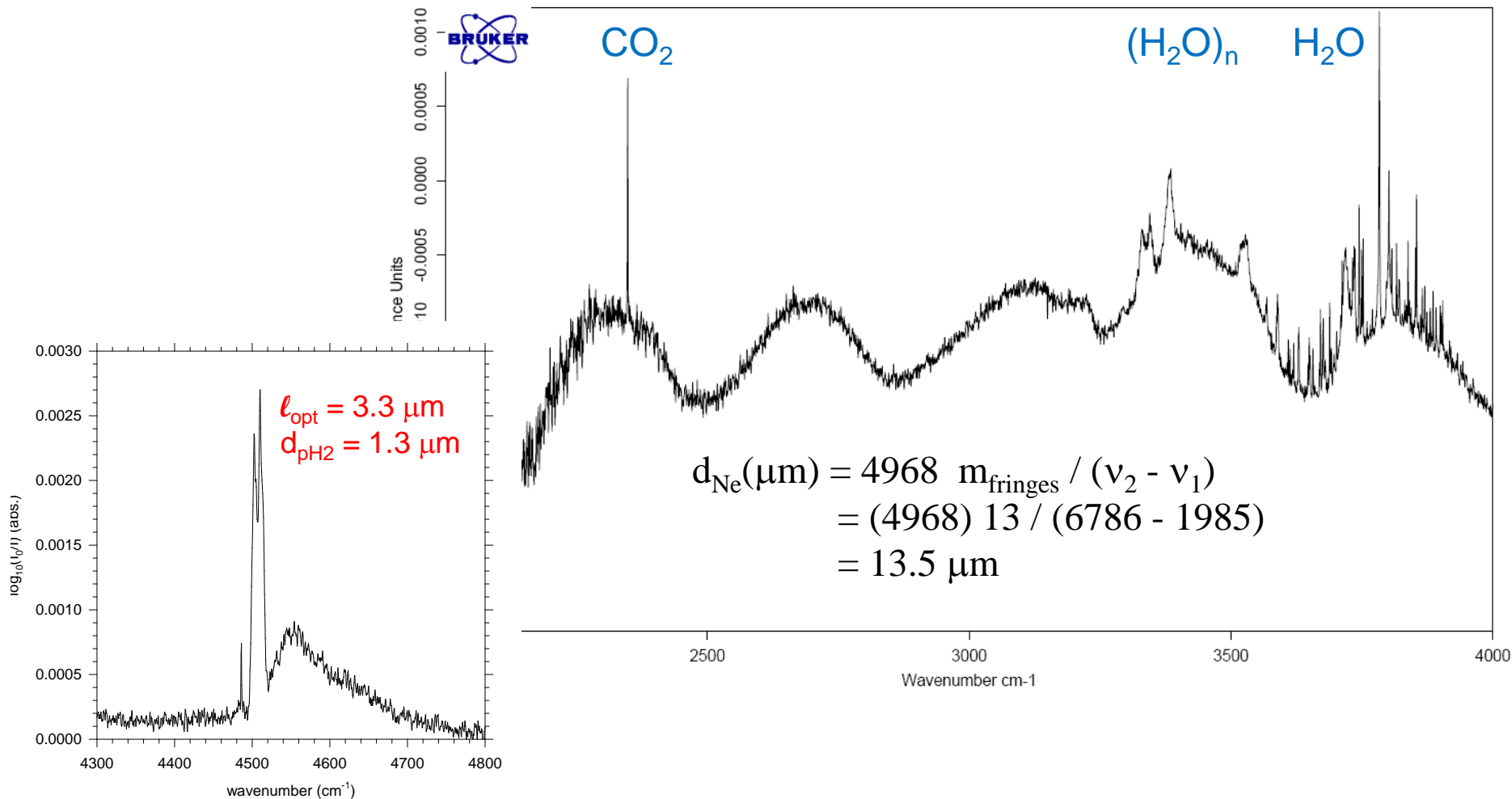
500 ppm oH₂/pH₂ Overcoat



Solid pH₂ appears to be a very poor positron moderator!



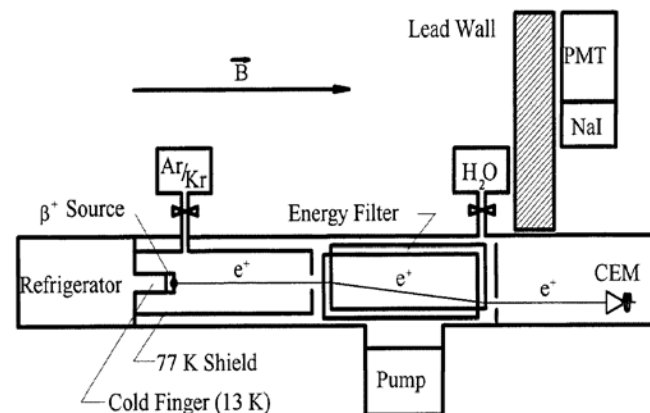
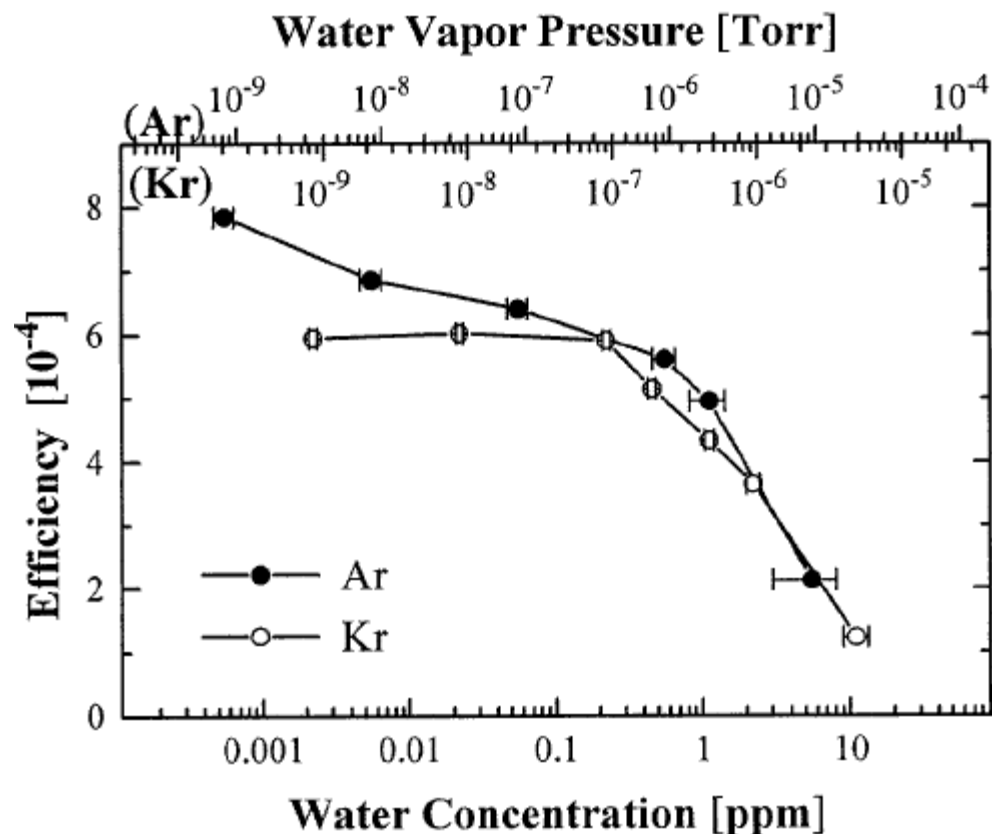
Dirty Ne Moderator + pH₂ Overcoat



Ne deposited from dopant manifold. $P_{\text{cryo}}(300\text{K}) = 1.1 \times 10^{-8}$ torr.



Effects of H₂O Impurities

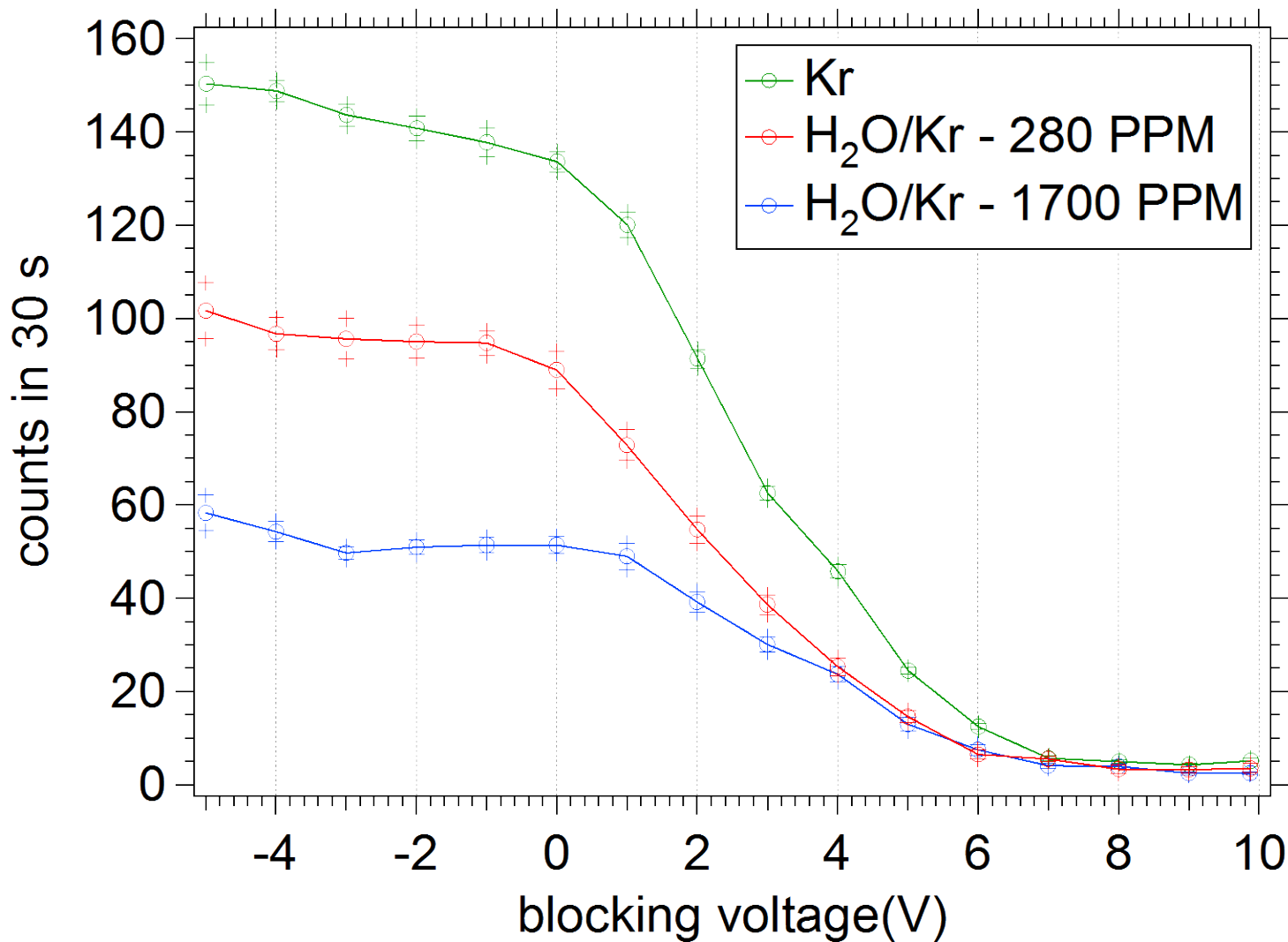


H₂O concentrations estimated from H₂O partial pressures in deposition chamber.

M.P. Petkov, K.G. Lynn, and L.O. Roellig, J. Phys. Cond. Mat. **8**, L611 (1996).

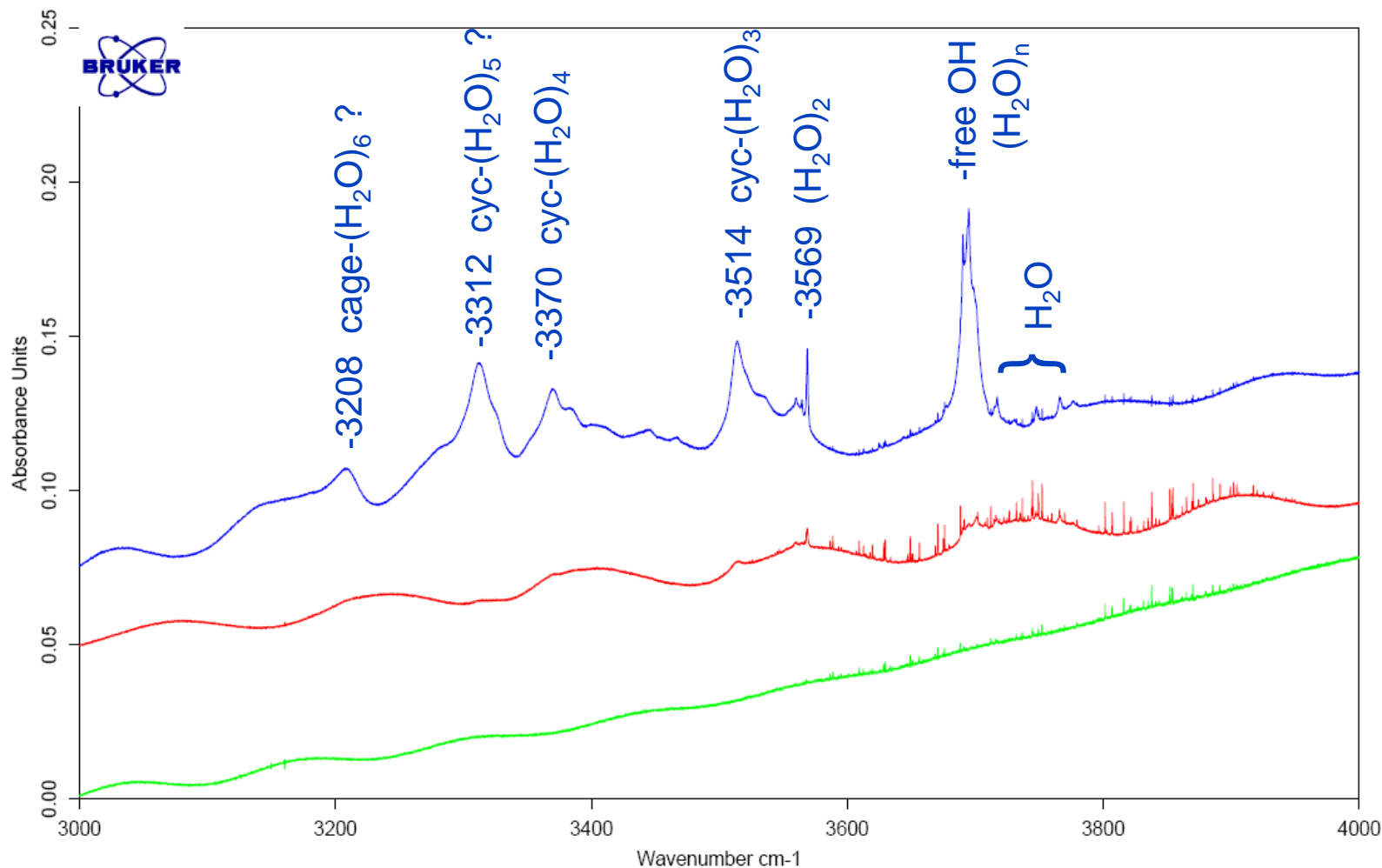


H₂O/Kr Moderators





H₂O/Kr Moderators



nominal
H₂O conc.
& thickness

1700 ppm
d = 33 μ m

280 ppm
d = 25 μ m

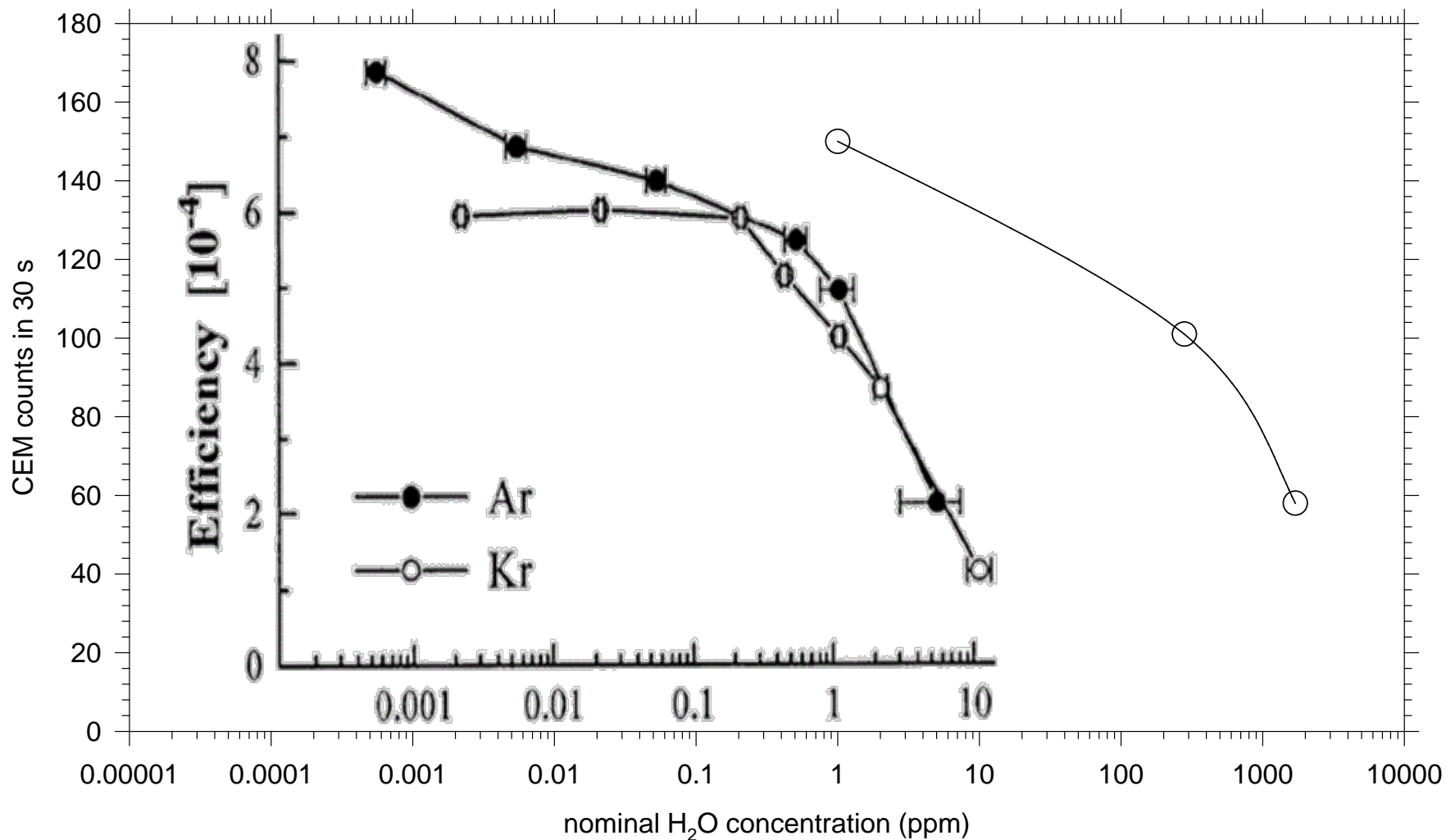
~1(<20) ppm
d = 31 μ m

(H₂O)_n/Kr assignments – A. Engdahl and B. Nelander, J. Mol. Struct. **193**, 101 (1989).

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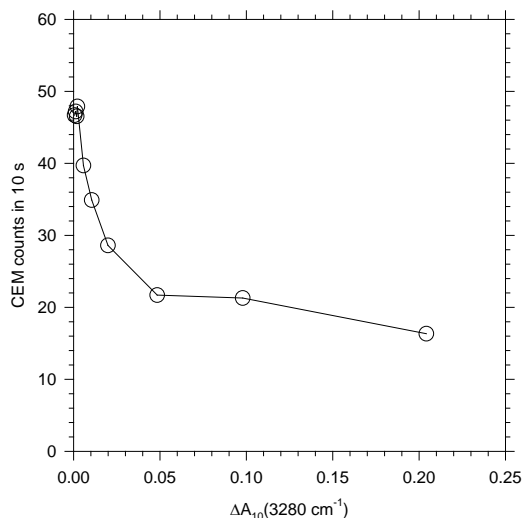
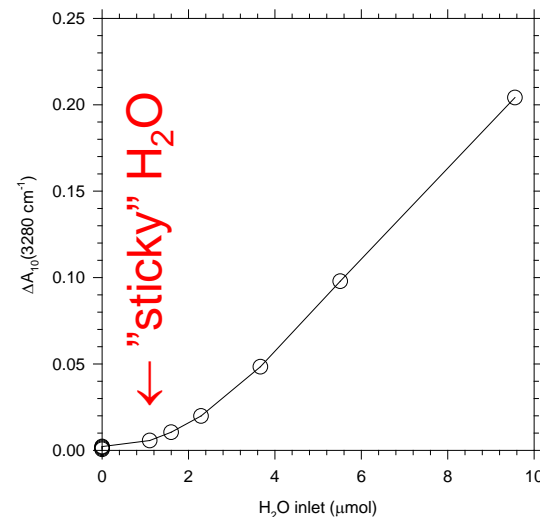
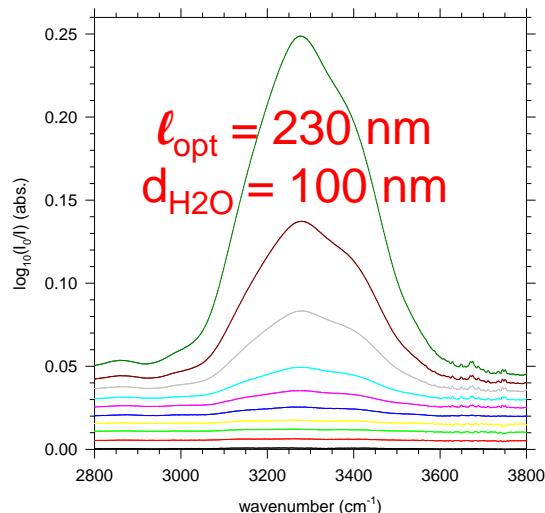
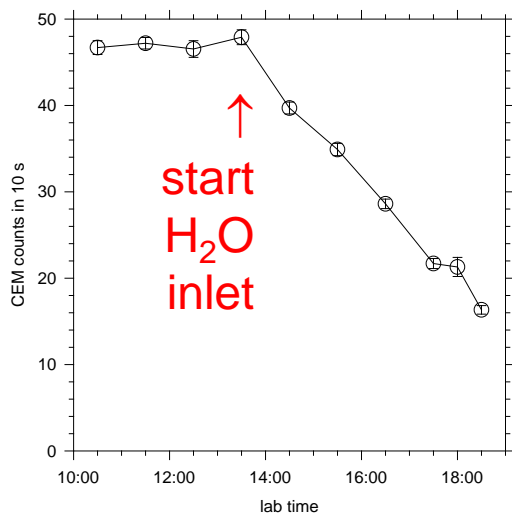
H₂O/Kr Moderators



H₂O concentration estimates differ by two orders of magnitude.



H₂O Ice Overcoat on Kr Moderator



FTIR spectrum consistent with amorphous ice, as expected for vapor deposition with $T_{\text{sub}} = 30 \text{ K}$.

[W. Hagen, A.G.G.M. Tielens, and J.M. Greenberg, CP **56**, 367 (1981).]

See initial rapid drop in slow e^+ yield, as expected.

Plateau in slow e^+ signal for thick ice not expected;
 \Rightarrow porous ice, possible experimental artifact (?!).



Summary & Future Directions



- Demonstrated an apparatus for combined positron moderation + matrix isolation spectroscopy; permits testing of theories proposed to explain observed moderator efficiency reductions upon extended operation.
- Unfortunately, ~ 500 ppm oH₂/pH₂ cryogenic solids are poor e⁺ moderators; $\epsilon_{\text{pH}_2} < 0.01 \epsilon_{\text{Ne}}$.
- Previous studies of the role of water impurities in Rg moderators may have seriously underestimated trapped H₂O concentrations.
- Slow positrons can emerge from Kr moderator coated with ≈ 100 nm amorphous water ice.
- One last set of experiments planned for the end of FY11. Will repair ortho/para hydrogen converter to bring residual oH₂ concentrations below 100 ppm.

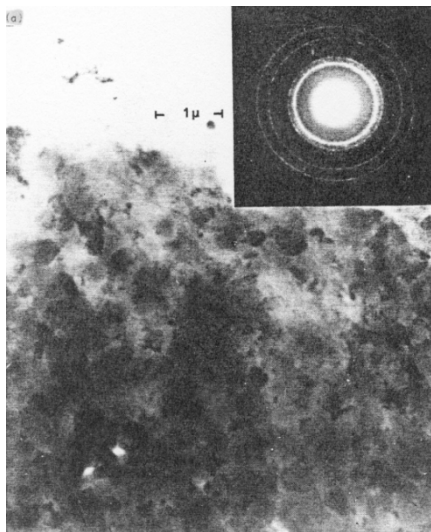


Backup Slides



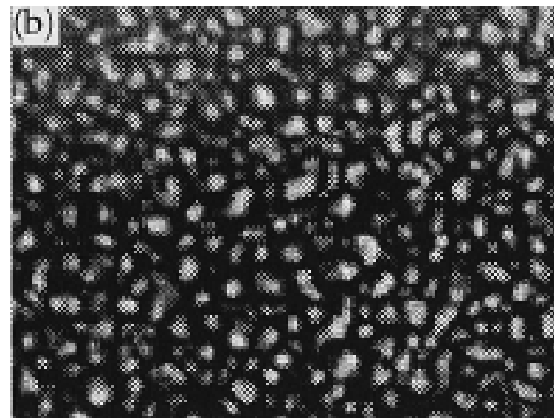


Solid Ne vs. Solid pH_2



Electron micrograph and diffraction pattern of vapor deposited solid Ne showing sub-micron scale defects.

J.A. Venables and B.L. Smith, "Crystal Growth and Crystal Defects," in Rare Gas Solids Vol. II, edited by M.L. Klein and J.A. Venables (Academic Press, London, 1977).



|----- 1.7 mm -----|

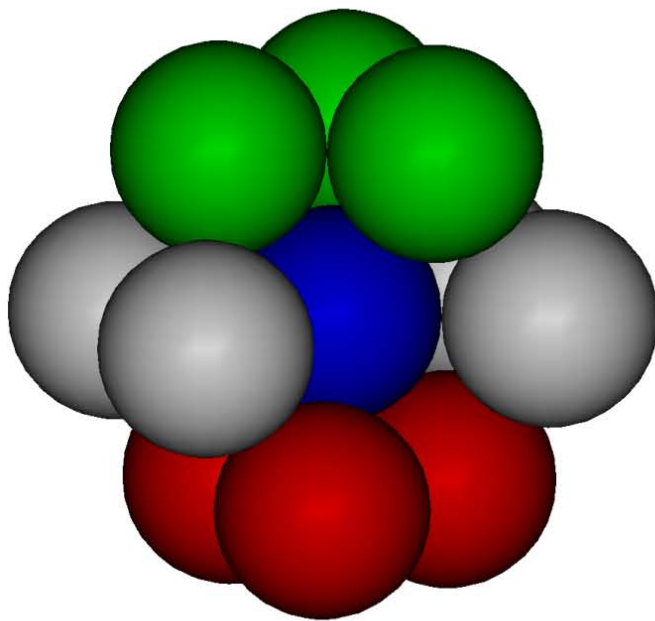
Typical crystallite size in vapor deposited pH_2 is $\sim 100 \mu m$, or $\sim 1000x$ larger than for vapor deposited Ne.

G.W. Collins, W.G. Unites, E.R. Mapoles, and T.P. Bernat, Phys. Rev. B **53**, 102 (1996).

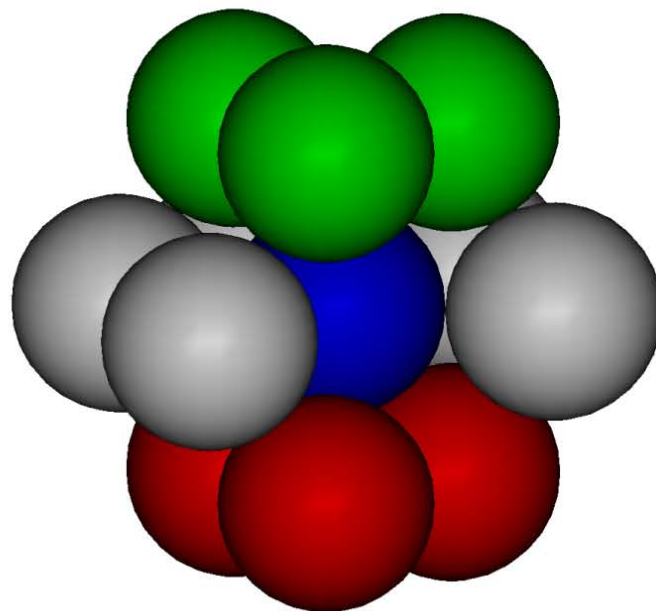
Also: solid pH_2 is a special material – a “quantum solid.”



fcc & hcp Structures



“face-centered cubic”
fcc \Rightarrow O_h site symmetry,
nearly isotropic

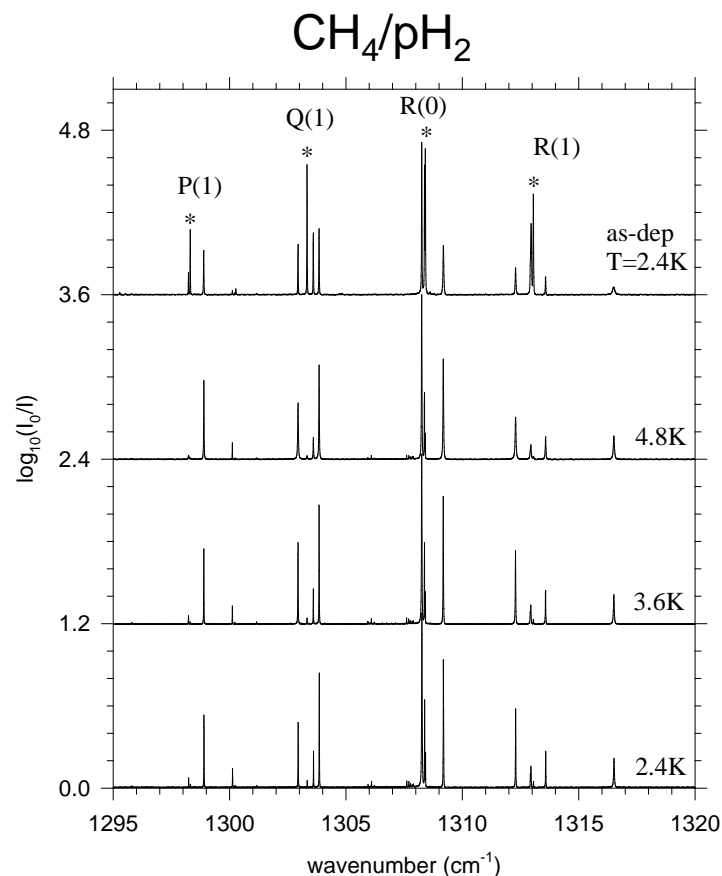


“hexagonal close-packed”
hcp \Rightarrow D_{3h} site symmetry,
unique c-axis

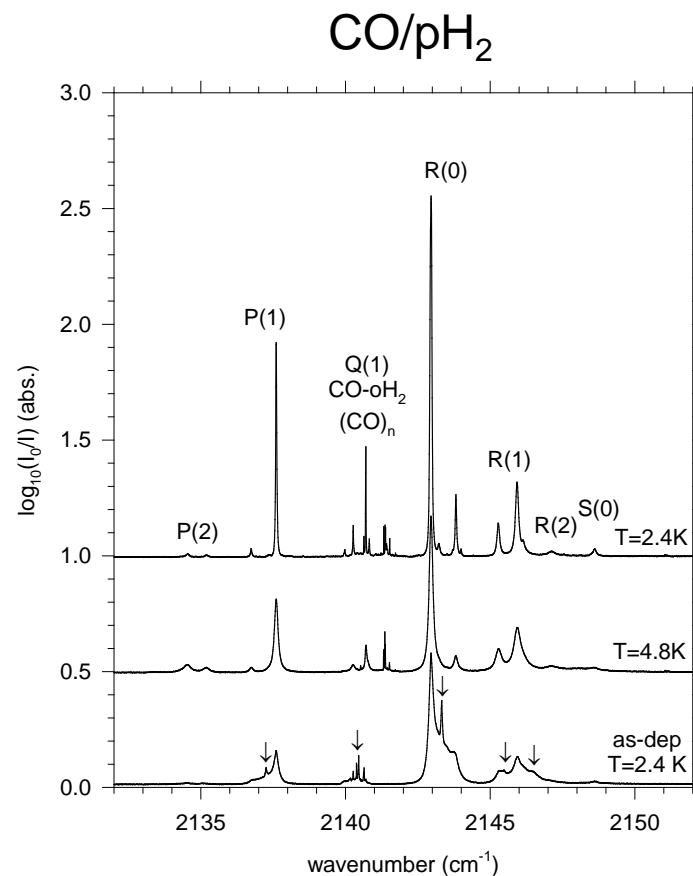
Use dopant molecules in solid pH_2 as probes to determine crystal structure.



fcc/hcp Regions in RVD Solid pH_2



S. Tam, M.E. Fajardo, H. Katsuki, H. Hoshina, T. Wakabayashi, and T. Momose, J. Chem. Phys. **111**, 4191 (1999).

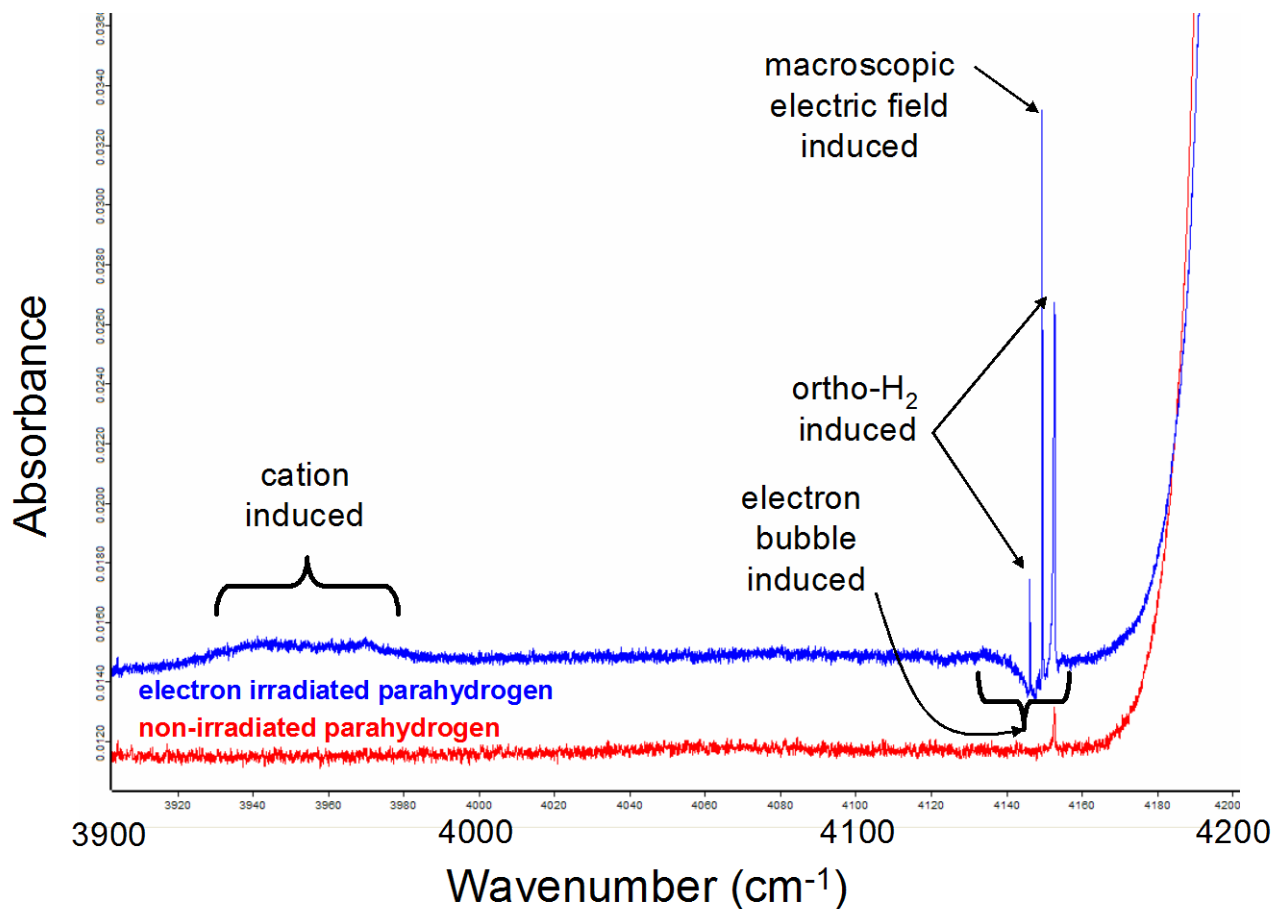


M.E. Fajardo, C.M. Lindsay, and T. Momose, J. Chem. Phys. **130**, 244508 (2009).

Mixed fcc/hcp microstructure in as-deposited samples; anneals to hcp.



Radiation-Induced Features

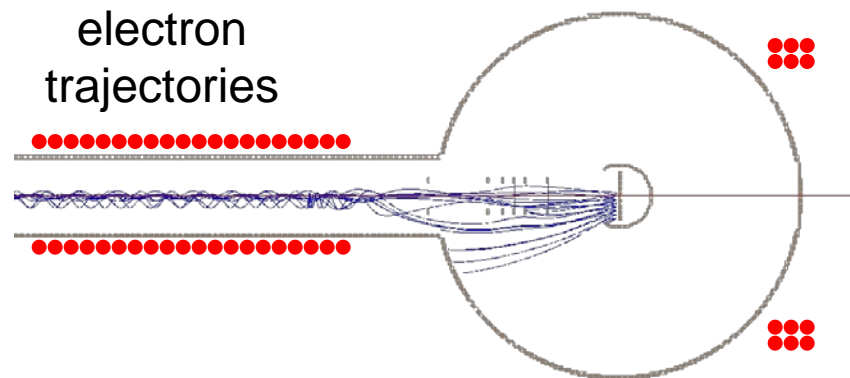
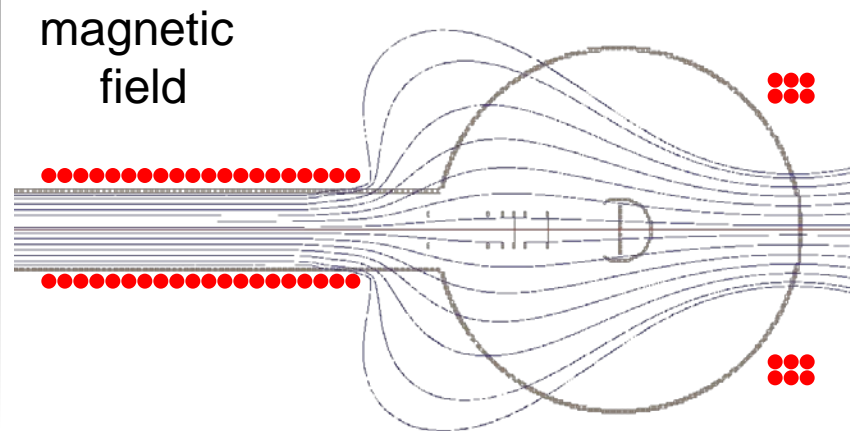
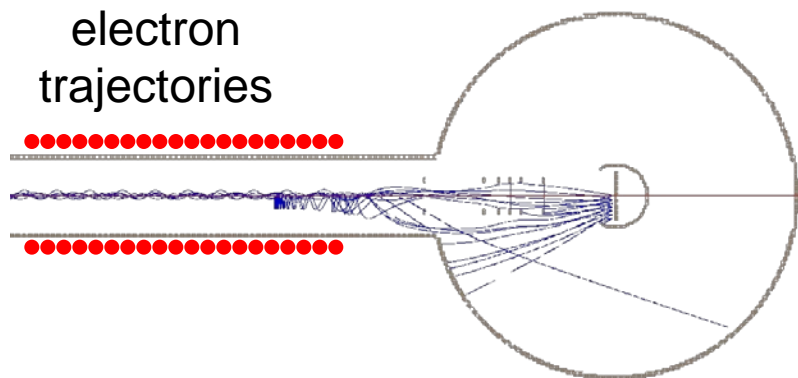
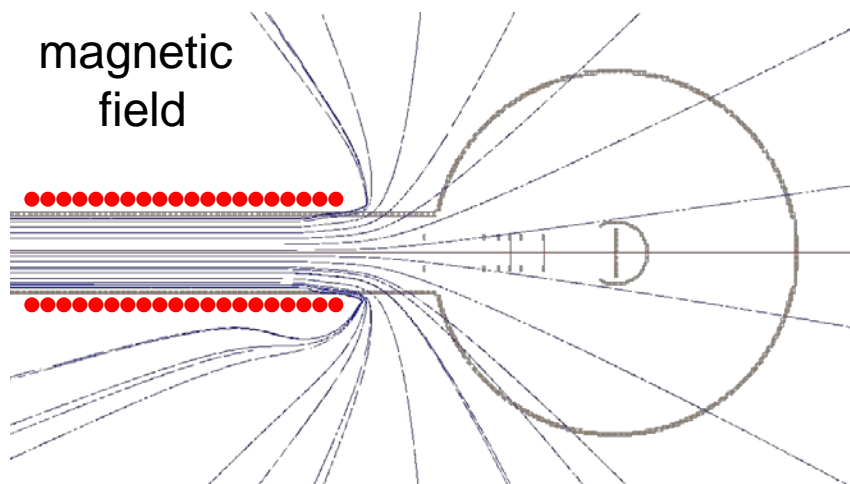


Monitor buildup of radiation damage *in-situ* during moderator operation.

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Modify Solenoid “Magnetic Funnel”



More electrons collected, but some trajectories evade energy selection gate!

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